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Novel Textile Actuator Having a Large Displacement Consisting of Electrospun Poly(DL-Lactic Acid) Fibrous Film and Its Mechanism

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Abstract

We found that a racemic poly(lactic acid) (PDLLA) fibrous film prepared by the electrospinning method has a unique spontaneous polarization and exhibits inverse-piezoelectric behavior. These properties were characterized by displacement monitoring and thermally stimulated current measurements.

1. Introduction

Poly(lactic acid) (PLA) is a biodegradable thermoplastic polyester. Moreover , it is commonly known that stretching to orient films of PLAs that exhibit chirality, i.e., poly(L-lactic acid) and poly(D-lactic acid), results in a shear piezoelectric effect [1,2]. On the other hand, PLA containing D- and L-lactide as a racemic mixture, i.e., poly(DL-lactic acid) (PDLLA), does not exhibit piezoelectricity. However, in this study, we found the unique actuation behavior of a randomly oriented electrospun fibrous PDLLA film [3]. The strain in the film caused by the application of a voltage was quite large: a strain of 9% at –100 V; thus, its actuation behavior will be employed in some applications such as textile actuators and buzzers for wearable devices. This actuation was attributed to an inverse-piezoelectric-like behavior, despite the nonpiezoelectric and racemic PLA content. Its polarization was also characterized by examining the current peaks in the thermally stimulated current (TSC) spectrum of the PDLLA film [4].

2. Measurements

2.1. Materials

PDLLA (Mw = 300,000–600,000) was purchased from Polyscience Inc. and used as-received. Chloroform (99.0%) and N,N-dimethylformamide (DMF) (99.5%) were purchased from Kanto Chemical Co., Inc., and also used as-received.

2.2. Electrospun fibers [5]

PDLLA was dissolved in a mixture of chloroform and DMF (chloroform:DMF = 3:2 by volume) at a concentration of 120 mg•ml^{-1}. The solution was then continuously supplied to a plate collector at a pump rate of 0.04 ml•h^{-1} using a glass syringe equipped with a stainless-steel needle (0.18 mm in diameter). A high-voltage power supply was connected to the needle at a voltage of 4.0 kV. The plate collector was a Ag electrode formed on a polyimide film using screen printing. The distance between the needle and the plate collector was 10 cm. Electrospun PDLLA fibers were continually deposited onto the collector for a period of 30 min. All experiments were performed in air at room temperature and a humidity of 50 ± 5% RH. As a reference sample, an amorphous PDLLA film comprising a nonfibrous structure was also prepared on a Ag electrode/polyimide substrate by spin coating at 500 rpm for 30 s using the same solution employed for electrospinning.

2.3. Characterization

The shapes of the fibers were characterized using field-emission scanning electron microscopy (FE-SEM). The mean diameter of the fibers was calculated from 270 SEM images of individual fibers. The displacement of the film surface was observed by a laser confocal displacement meter with a voltage applied to the film by a source/measure unit. The voltage was applied to the bottom electrode on the polyimide substrate of the sample with its top electrode grounded. The displacement was measured at the center of the film surface over the top electrode. For TSC measurements, both electrospun and spin-coated samples were first cooled to –120 °C at a rate of 20 °C/min and then heated to a temperature of 60 °C at a rate of 10 °C/min in a chamber filled with He gas. In these measurements, a poling treatment was not performed; therefore, only the spontaneous polarization of the samples was observed. The thermal relaxation current of the spontaneous polarization was measured while increasing the sample temperature.

3. Results and Discussion

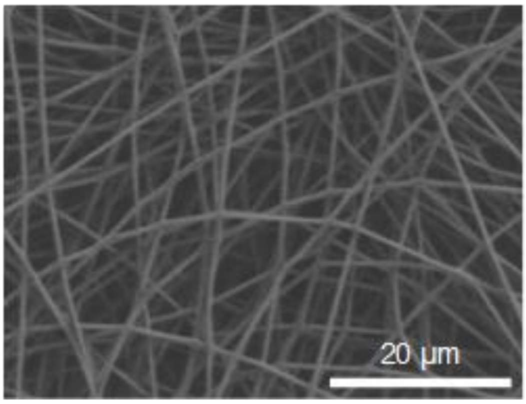

Figure 1. FE-SEM image of the surface of an electrospun PDLLA fibrous film.

Figure 1 shows FE-SEM images of the surface of an electrospun PDLLA-fibrous film. The average diameter of the PDLLA fibers in this film was estimated to be 730 nm. This film consists of many stacked PDLLA fibers with air pockets between them. The density of the PDLLA fibers in this film can be estimated using the thickness and relative permittivity of the film, and the calculated density was ~20%. From its density, this electrospun PDLLA-fibrous film had a particularly spongy structure. We expected that this sponge-like fibrous film would act as a dielectric elastomer that contracts in the thickness direction owing to an applied electrostatic pressure.

To demonstrate the actuation of a PDLLA-fibrous film, a change in the thickness of the film was observed. Figure 2 shows the displacement of the film surface and the displacement current when a triangular wave with an amplitude of ±100 VPP was applied to the film as the applied voltage. Interestingly, the surface displacement shows an essentially linear variation that is similar to the inverse-piezoelectric effect of piezoelectric materials. The piezoelectric-like behavior of this PDLLA-fibrous film is very unique because PDLLA as a D/L random polymer should not generally exhibit piezoelectricity.

We thought that the electrospun fibrous PDLLA film had a unique spontaneous polarization that resulted in piezoelectric-like effect. Thus, TSC measurements were obtained for the film. If the film exhibited a spontaneous polarization, TSC peaks should appear in the absence of a poling treatment. The TSC spectra of the electrospun PDLLA film (A) and the spin-coated PDLLA film (B, as a reference of (A)) are shown in Figure 3. In the case of the electrospun sample, there were negative peaks at around –35 °C and 0 °C. However, these peaks were not observed in the case of the spin-coated film. These TSC results indicate that the electrospun PDLLA fibrous film exhibited a polarization, even though a poling treatment had not been conducted. When the spontaneous polarization was thermally relaxed, charge compensating for the polarization was detected. We propose that the PDLLA film exhibited a spontaneous polarization as a result of the strong electric field applied to the PDLLA polymer solution used in the electrospinning approach.

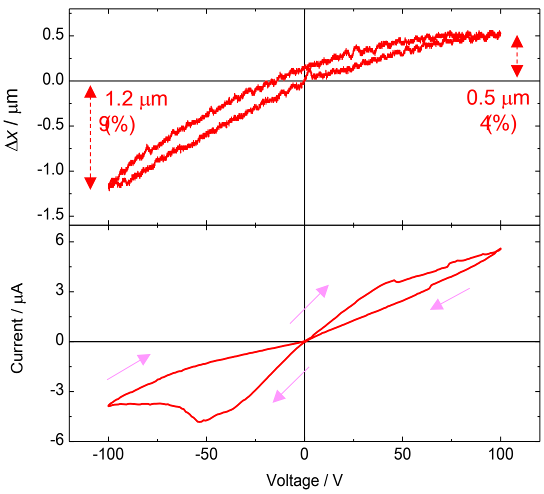

Figure 2. Displacement of the surface of an electrospun PLA-fiber film and the displacement current flowing in the film versus the applied voltage—a triangular wave with a scan rate of 6 V/s and amplitudes of ±100 V_{PP}.

4. Conclusions

The electrospun PDLLA-fibrous film had a spongy structure with a 20% density of PDLLA fibers in the film. The fiber film exhibited expansion and compression under the application of a voltage. Its actuation mechanism is likely caused by a piezoelectric-like variation in the film thickness. Furthermore, a large strain of 9% was induced in the thickness direction when a potential of –100 V was applied to such a film. TSC measurements were also conducted to demonstrate that electrospun fibrous PDLLA films exhibit a spontaneous polarization owing to the presence of a strong electric field applied to the PDLLA solution while forming the fibers. The unique polarization characteristics exhibited by the electrospun PDLLA film appeared to contribute to its piezoelectric-like properties.

References

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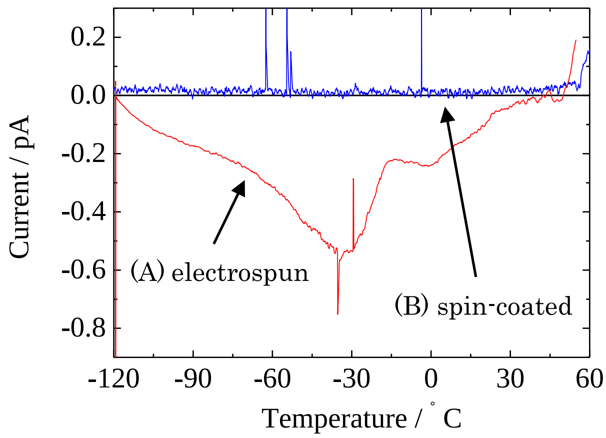

Figure 3. TSC spectra for (A) an electrospun film and (B) a spin-coated film of PDLLA without a poling treatment. The scanning temperature range was in the range of -120–60 °C with a heating rate of 10 °C/min.