Large, Fast, and Bidirectional Bending of Slide-Ring Polymer Materials

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The bending of slide-ring materials (SRMs), polymer materials having ring-shaped cross-linking points providing deterioration tolerance and low hysteresis, is described. An SRM sheet exhibits a voltage-controllable, extremely large, fast, and bidirectional bending deformation with shape memory functionality. A 50-µm-thick SRM sheet actuator exhibits an electrostatic actuation in two opposite directions and bending angle larger than 180° achieved within 0.4 s due to its shape memory effect. The memory effect remains for more than 250 min. As a potential application utilizing the bending of the SRM, a soft gripper is fabricated, which handles two objects placed opposite each other. This functionality is rarely achieved by a soft gripper composed of a single active material, which shows the characteristics of the SRM promising for the development of soft matter systems with novel functionalities and high performances.

The animals utilize their bodies and fingers composed of soft structures such as tissue and skin in various activities. For use in robots, the inherent structural compliance improves the absorption of mechanical impacts,[1,2] enables the gripping of fragile and sensitive objects,[3,4] and provides versatility and simpler mechanical design and control input.[5–7] These characteristics are often difficult to achieve by conventional robotics using rigid materials. Thus, extensive studies have been carried out on the development of soft robots using compliant materials for various applications, from simple soft grippers to dexterous robotic systems, such as multimodal locomotion robots and rehabilitation devices.[6,8–11] Multi-functional active materials having actuation capabilities are essential for next-generation soft-matter systems.[6,7,12,13] In particular, the bending actuation is the most versatile actuation type for diverse applications.[14] Various soft active materials exist.[12,14] Among them, the fluidic elastomer actuators (FEAs)[11,13] are widely used. The FEAs operate by injecting a fluid such as air and dielectric liquid. They have high design flexibilities and can output large actuation strokes and forces. However, the need for pump or compressor leads to a heavy and bulky structure, which often limits the mobilities of soft robots. Electroactive polymers (EAPs),[16–18] which deform under electrical stimuli, are another candidates for soft robotic multi-functional materials. Mainly two types of EAPs exist, ionic polymer–metal composites (IPMCs) and dielectric elastomer actuators (DEAs). The IPMCs are composed of a fluorine-based ion-exchange resin sheet laminated between two electrodes. When a voltage is applied, positively charged cations in the composite move to the negative side, which leads to a bending of the sheet toward the positive side. The IPMCs can generate large bidirectional bending actuations at low driving voltages (few volts), but their actuation speeds are generally low (e.g., 3.5 min to achieve a bending angle of ≈270°) and often the actuator structure needs to be wet. In contrast, the DEAs can operate under dry conditions. They consist of an elastomeric sheet with compliant electrodes on both sides. A high applied voltage (few kilovolts) squeezes the sheet due to the Maxwell stress induced between the two electrodes, which expands the area. The DEAs exhibit large actuation strains (e.g., larger than 100%)[20,21] and fast responses. Combined with an additional substrate, they can even produce bending deformation.[17,22] However, the bending actuation is limited to only one direction. The soft actuator technologies discussed above suggest the lack of active soft materials capable of generating a large fast bidirectional bending actuation with simple structure and input source.

In this article, we report an unprecedented bending of a thin elastomeric sheet, which could address the current limitation of multi-functional materials and pave the way for next-generation soft robotics. The used elastic sheet is a type of polymer, which could exhibit bidirectional extremely large bending deformations.
electrostatically induced as in the DEAs. However, unlike the DEAs, our polymeric actuator could reverse the bending direction only by inverting the applied voltage. Moreover, we demonstrate memory-like characteristics, i.e., the sheet recovers to the previous bending displacement quickly upon voltage application after some nonactivation time. The sheet-like structure provides a high design flexibility, which enables the fabrication of devices with various geometries.

A slide-ring material (SRM) is used as the polymer material in this study. Such polymer compounds have been reported for the first time by Ito et al.[23] The SRMs are fabricated using rotaxane, in which each polymer chain penetrates ring molecules. The ring molecules can move through the polymer chains. This characteristic, referred to as pulley effect, provides an excellent hysteresis behavior.

In this study, a thin SRM sheet exhibits bending under an electric field. The configuration of the sheet is the same as that of the DEA, i.e., a thin SRM sheet is sandwiched between two compliant electrodes. However, the actuation is quite different. The SRMs can generate bending actuation without additional element, whereas the DEAs require additional substrates to achieve the same actuation. Therefore, as an active material capable of outputting practical deformation, the SRM has a considerably simpler structure enabling a high design flexibility and small weight.

Bending of SRMs has not been previously reported. However, a similar behavior has been observed in polyurethane sheets.[24] This phenomenon is referred to as bending electrostriction, which has many similarities to the bending actuation of the SRMs. The thin polyurethane sheet with compliant electrodes on both sides has exhibited a transition of bending displacement and memory effect similar to those of the SRMs, although the displacement has been considerably smaller.[24] The mechanism of the bending of the SRMs has not been revealed. We assume that it is attributed to the same mechanism as in the bending electrostriction of polyurethane.

We illustrate the bending of the SRM in Figure 1. When a high voltage is applied to the electrodes, electric charges are injected inside the SRM sheet. The injected charges are referred to as space charge. The space charge leads to a nonuniform electric field in the elastomer sheet. Thus, a biased internal stress appears, leading to the bending of the sheet. In other words, the part of the sheet between the space charge and opposing charges in the electrode (lower half in Figure 1b) functions as an expanding active layer, whereas the rest (upper half in Figure 1b) acts as an inactive layer, generating a bending deformation in a manner similar to that in a unimorph actuator. The resulting shape of the bending deformation depends on the electrical and mechanical conditions. The inversion of the polarity of the applied voltage leads to bending in the reverse direction. In this case, the distribution of space charge is opposite to that in the case in Figure 1. The space charge distribution can be measured using the pulsed electroacoustic method.[25] In the case of polyurethane bending electrostriction, the internal space charge distribution was measured using this method and the actual bending was calculated using the measured space charge distribution.[26] The memory effect of the SRM, as a part of the bending phenomenon, can be explained by the presence of space charge. Once the charge injection occurs, the formed space charges remain for a certain period in the sheet and their distribution does not quickly change.

To elucidate the bending of the SRM and its electromechanical behavior, we assessed the bending characteristics of an SRM sheet actuator. A 50-μm-thick SRM sheet was cut to a triangular shape. Carbon black (Ketjenblack EC-300J) was used to form the electrodes. The SRM sheet was initially slightly bent due to the residual stress originated from the fabrication. The heat curing caused the thermal expansion of the material, which led to a nonuniform remaining stress along the thickness direction. The curvature of the bottom part of the triangular actuator was measured as a function of the applied voltage using a line

Figure 1. Bending of the SRM. a) The bending actuation is achieved by a configuration consisting of an SRM sheet and two compliant electrodes. b) When a high voltage is applied to the electrodes, charge injection occurs and leads to space charge generated at a biased position along the thickness direction. The space charge leads to asymmetric internal stress distribution and bending stress, and thus to a large bending deformation. Bending in the opposite direction occurs upon the inversion of the polarity of the applied voltage.
laser shape meter (LS-100CN, OPTEX FA). We applied a potential difference up to 1000 V to the actuator, because the maximum excitation voltage (i.e., breakdown voltage), which differs among samples, was typically approximately 2000 V. The measured voltage–curvature transition is shown in Figure 2a. Before the experiment, the SRM sheet did not experience voltage application. In the range of 0 to 10 s, voltage was not applied and the curvature was 0.1 mm⁻¹ because of the residual stress. In the range of 10 to 70 s, a direct-current (DC) voltage of 500 V was applied to the sheet. The curvature gradually increased and saturated at 0.17 mm⁻¹. The increase in curvature implies that the sheet exhibited bending deformation. The curvature of 0.17 mm⁻¹ corresponds to a bending angle of 195°. The observed transition of the curvature reflects the charge injection, which accumulated space charge until the actuator reached an equilibrium state. At 70 s, once the voltage application was terminated, the actuator immediately returned to its initial bending state (response time ≈0.4 s). The voltage was applied again at 80 s and the actuator quickly restored the curvature (response time ≈0.4 s) due to the shape memory effect. The second bending was considerably faster than the first bending, because of the space charge in the SRM sheet, which functioned as a memory storing the deformation.

Figure 2b shows the bending characteristics of the SRM actuator under inverted voltages. The bendings to the front and back sides correspond to positive and negative values of curvature, respectively. The insets above the graph represent the actual shapes of the SRM sheet actuator during the measurement. The initial curvature had a positive value (0.1 mm⁻¹), according to the data at 0–10 s. In the range of 10–40 s, the actuator bent as in the previous experiment. The voltage application was terminated in the range of 40–50 s, and then an inverted voltage was applied (−750 V). Immediately after the voltage application, the actuator quickly bent to the same direction as before, but the curvature gradually decreased and it became flat at 60 s. The spikes around 60 s are attributed to the instability of the curvature calculation for a flat surface. Subsequently, the actuator started to bend toward the opposite direction. The transition of the deformation was similar to that in the case of the application of the positive voltage; the absolute value of the curvature gradually increased. Before cutting off the voltage at 110 s, the curvature reached −0.15 mm⁻¹. In the rest state, the actuator had a curvature of 0.08 mm⁻¹.

As demonstrated experimentally, in the bending of the SRM, the sheet reached an equilibrium state after a certain time period (i.e., above a certain amount of charge injection). We explain the bending characteristics of the SRM at different applied voltages. In this experiment, the line laser shape meter was not used because the sheet became completely rounded at high voltages due to the extremely large bending and the curvature could

Figure 2. a) Response of the SRM bending actuator under a constant applied voltage. b) Bending characteristics of the SRM actuator under inverse applied voltages. The sign of the data in the range of 60–110 s is reversed to simply illustrate the deformation behavior. c) Bending actuations for both positive and negative voltage potentials (up to ±1000 V). d) Time constant of the bending actuation as a function of the voltage-off time. The shape memory effect remains for more than 250 min.
not be measured using the shape meter. Instead, we captured the actuator shape from the bottom by a camera. Subsequently, the curvature was obtained by image processing. Figure 2c shows the relation between the applied voltage and curvature in the steady state for each voltage. For both positive and negative voltages (up to $\pm 1000 \text{ V}$), the actuator exhibited a quadratic dependence, because the Maxwell stress is proportional to the square of the electric field. The maximum measured curvatures were $0.33 \text{ mm}^{-1}$ at $1000 \text{ V}$ and $0.08 \text{ mm}^{-1}$ at $-1000 \text{ V}$, corresponding to bending angles of $378^\circ$ and $91^\circ$, respectively. This demonstrates that the large bidirectional actuation of the SRM can be controlled by the applied voltage. We also attempted to measure the output force of the SRM actuator. However, because of the softness and thinness of the SRM sheet, the force was extremely small so that it could not be measured by our experimental setup.

Further, we characterized the memory effect of the SRM sheet actuator to identify its duration. First, the voltage application was terminated for a certain period after reaching a steady state under a constant voltage. Subsequently, the voltage was applied again to investigate the duration of the memory effect. The input voltage was $750 \text{ V DC}$ in this experiment. The memory effect was evaluated using the time constant to reach $63.2\%$ of the total displacement, based on the curvature at $45 \text{ s}$ after the voltage application. Figure 2d shows the data obtained through the measurement. The time constant remained small ($\approx 4 \text{ s}$) for rest times smaller than $250 \text{ min}$, which implies that the memory effect existed for this time duration. After $500 \text{ min}$, the time constant suddenly increased ($\approx 26 \text{ s}$), which suggests that the memory effect disappeared at a critical point in the range of $250$–$500 \text{ min}$.

One of the potential applications utilizing the bending of the SRM is soft grippers capable of bidirectional grasping. Figure 3 shows a prototype of a soft gripper composed of a single SRM layer. In addition to the physical actuated grasping, the device involves electroadhesion, which improves the shear force to lift up the object. The electroadhesion has a good compatibility with SRM-based electrostatic devices because the driving voltage is at the same level in both technologies. In the gripper, the upper part provides bending actuation, whereas the lower part wraps the object and generates electroadhesion by the interdigital electrodes. The gripper weighs only $0.3 \text{ g}$, but it can grasp and lift up an object with a mass of $4.4 \text{ g}$, more than ten times its mass. In the sequence of operations in Figure 3, the objects are composed of acryl square poll with paper on the surface. Due to the large bidirectional bending actuation, the gripper successfully manipulated the two objects placed opposite each other.

We have revealed an unprecedented behavior of the SRM, which has not been observed in other polymeric active materials. The SRM sheet actuator exhibited a voltage-controllable extremely large fast bidirectional bending actuation to a bending angle larger than $180^\circ$ in $\approx 0.4 \text{ s}$. The actuator generated the bending deformation without additional part, which is the main structural difference from other DEA-based bending actuators, which require a passive element such as a stretchable substrate or inextensible layer. This characteristic makes our actuator simple and ensures a high design flexibility. Moreover, unlike the existing DEAs, our device could bend in the direction perpendicular to the longitudinal direction due to the triangular shape. The fast response of the actuator is associated with the shape memory, which remembers the previous actuated deformation for more than $250 \text{ min}$. The developed gripper was used to manipulate two objects placed opposite each other by utilizing the large bidirectional bending actuation. This is an unusual functionality rarely achieved by soft grippers composed of a single active material, which simplifies the handling of several objects. The output force of the SRM actuator is an important factor determining the electromechanical behavior, which will be investigated in our future study. The characteristics of the SRM analyzed in this study make it a promising active material for the development of soft-matter systems with novel

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**Figure 3.** Soft gripper composed of the SRM. The device utilizes the bending of the SRM combined with the electroadhesion generated by the interdigitated electrodes to improve the lifting force. The gripper can bend its finger bidirectionally, which enables selective gripping of the objects placed opposite each other.
functionalities and high performances for various applications including soft robotics, wearables, haptics, and industry applications. Particularly, microdevices are expected to benefit from the simplified structures of the SRM actuators. Further optimization of the material in terms of composition and structural design is required to match the performances of the devices discussed previously.

Experimental Section

Fabrication of the Triangular Actuator and Gripper: The triangular actuator and gripper in this study were fabricated using SRM sheets with the same composition and same electrode material. The SRM sheets were fabricated by solvent casting (thickness: 50 µm) and exhibited nominal Young’s moduli of 3.4 MPa. In this method, a polymer solution consisting of polypropylene glycol and grafted polyrotaxanes having α-cyclodextrins with poly-ε-caprolactone chains (Advanced Soft Materials Inc.) was casted on a film substrate and cured in an oven. The sheets were produced in-house by the groups of one of the authors. Carbon black (Ketjenblack EC-300D) was used for the electrodes because of the good bending characteristics and simple handling. After the devices were cut out from an SRM sheet, electrodes were patterned using a brush and mask of a polyethylene terephthalate sheet. A conductive tape was used to obtain electrical connection. The electrode dimensions of the triangular actuator were 18 mm (width) × 25 mm (length), whereas the total width of the actuator was 20 mm. The electrode width of the gripper was 40 mm, whereas the length was 55 mm.

Experimental Setup for Actuator Characterization and Gripper Demonstration: The bending of the SRM sheet was measured using a line laser shape meter (OPTEX FA, LS-100CN) with a sampling rate of 10 Hz. A voltage amplifier (TREK, 609C-6) and function generator (HIOKI, 7075) were used to drive the actuators and gripper. The curvature was calculated using the measured shape data for the central part (width: 6 mm) of the sheet and circular fitting.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

electroactive polymers, slide ring materials, soft actuators, soft grippers, soft robotics

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