Stretchable Light-Emitting Device Using a Film/Elastomer Bilayer System with Electrodes Patterned by Printed Electronics Technique

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Stretchable light-emitting sources have attracted attention for various applications such as deformable displays and health care devices that can be attached on non-flat shapes such as the human body. One conventional method of making stretchable devices is by designing stretchable wavy structures by attaching thin functional films on pre-stretched elastomers. However, this method requires a special apparatus to stretch the elastomer because of which printed electronics cannot be applied for film coating and patterning. In this study, we proposed a method that utilizes a plastic deformation, stretchable wrinkle structure consisting of conductive polymer electrodes which are patterned using printed electronics technique. Here, we present the fabrication method and the experimental results of the proposed material. Using this method, stretchable comb-shape electrodes were patterned on an elastomer/electroluminescent powder composite, and a light-emitting device stretchable up to an equi-biaxial stretch \( \lambda = 1.4 \) was fabricated. Our method will help in the fabrication of stretchable light sources.

Keywords: Stretchable, Light-emitting device, Printed electronics, Wrinkle, Plastic

1. Introduction

Light-emitting stretchable devices that are attachable onto three-dimensional surfaces have been studied for deformable displays [1,2] and biomedical equipment [3,4]. For making stretchable devices, buckling of thin film/elastomer bilayer systems has attracted attention. If a thin film is attached onto a pre-stretched elastomer and the stretch is released, a periodical buckling structure, also called a wrinkle structure, is formed on the thin film because of the gap in length between the film and the elastomer. If re-stretched, the wrinkle structure flattens and avoids fracturing.

This buckling phenomenon is multi-scale (in the order of sub-micrometers [5] to centimeters [6,7]) and is independent of the material (e.g., polymer [8], metal [9], oxidized elastomer [10,11], graphene [12], and skin [6,7]). For practical use, Rogers group at Illinois University attached a thin silicon device onto a pre-stretched elastomer and achieved a stretchable electronic device with a wrinkle structure [13]. However, for bonding a thin silicon device to an elastomer, complicated fabrication steps are required, such as precise adhesion configuration between the thin film and the elastomer. As an alternative, we previously presented a wrinkle-structured stretchable device utilizing plastic deformation of the conductive polymer [14]. Instead of attaching the thin film onto the pre-stretched elastomer, a plastic-conductive polymer was spin-coated on a non-stretched elastomer. By applying a stretching force to the plastic film/elastomer bilayer system and releasing the stretch, the elongated plastic film did not recover its initial length, and the length gap between the film and the elastomer was induced. As a result, a wrinkle structure emerged on the plastic-conductive film, and a stretchable conductive layer was achieved. The stretchable conductive layer was deposited on the surface of both sides of a functionalized elastomer that emits light under an electric field, and a stretchable light-emitting device
was achieved. With this method, stretchable devices can be made available in a simple manner.

Here, we present a more simplified method of fabricating stretchable light-emitting devices based on our previously proposed method. As presented in Fig. 1, comb-shape electrodes of plastic-conductive material were patterned on the top surface of a functionalized elastomer with the printed electronics technique. By applying a voltage to the comb-shape electrodes, the functionalized elastomer emitted light induced by the electric field between the electrodes. By simply applying and releasing the stretch, the wrinkle structure of the comb-shape electrodes was formed, and a stretchable light-emitting device was achieved. In this manner, electrodes patterned only on the top surface of the functionalized elastomer were sufficient to fabricate a stretchable light-emitting device. In addition, because no stretcher is required

Fig. 1. Schematic drawings of the proposed stretchable light emitting device. By applying a biaxial stretch and releasing the stretch, a two-dimensional wrinkle structure is formed on the surface. Under re-stretching, the wrinkle structure flattens and the device avoids fracturing.

Fig. 2. (a) Schematic drawings of the fabrication steps. (a1) A PDMS/EL composite was coated on a glass substrate. (a2) A parylene layer was coated as an adhesive layer. (a3) PEDOT:PSS electrodes were patterned on the top surface. (a4) Another parylene layer was coated as a protective layer. (b) A SEM image of the used cliché. (c) Schematic diagram of the electrode patterning with reverse offset printing. (c1) PEDOT:PSS was coated on a PDMS sheet. (c2) Electrodes were patterned on the PDMS sheet by partially transferring the PEDOT:PSS layer to the cliché. The electrodes patterned on the PDMS sheet were transferred to the structure depicted in (a2), and the structure depicted in (a3) is fabricated. (d) Photograph of the printed electrode patterns (gray patterns are PEDOT:PSS). (e) Smoothness evaluations of printed electrode pattern. (e1) Zoomed photograph, (e2) surface profile, and (e3) cross-sectional profile of the printed electrode patterns along the dashed line A-A' in (e1).
during the patterning process, the printed electronics technique is compatible with this method. In this study, we present the detail of the fabrication process and the resulting device.

2. Experimental

Figure 2(a) presents a schematic diagram of the fabrication process of our proposed light-emitting device. The fabrication process is partially followed by that reported in ref [14]. First, a liquid elastomer was prepared by mixing a base and a catalyst (weight ratio of base and catalyst = 5:1) of a commercially available elastomer kit (Sylgard 108, Dow Corning, Corp., Michigan, USA). Then, an electroluminescent (EL) powder (728 green EL phosphor, OSRAM SYLVANIA, Pennsylvania, USA: from the datasheet, the brightness of a screen-printed lamp using this material is 90 cd/m² at 100 V, 400 Hz) was mixed with the liquid elastomer (weight ratio, liquid elastomer: EL powder = 5:1). This functionalized elastomer was spin-coated on a glass substrate and cured at 75 ℃ for 2 h (Fig. 2(a1)). The thickness of the cured functionalized elastomer was approximately 300 μm. To fabricate the adhesive layer between the elastomer and the conductive layer, a layer of Parylene C (plastic material, Specialty Coating Systems, Inc., Indiana, USA) 0.7 μm thick was deposited on the functionalized EL elastomer (Fig. 2(a2)). Then, comb-shape electrodes were patterned (Fig. 2(a3)) with reverse offset printing as is briefly explained later. Finally, another Parylene layer 2 μm thick was coated on top of the sample as a protective layer (Fig. 2(a4)) to enhance the stretchability and durability of the electrodes [14].

Figures 2(b) and 2(c) present a cliché for reverse offset printing and the method of patterning the electrodes onto the sample. First, a cliché was prepared using a 4” silicon wafer 525 μm thick. AZP 4210 (Photoresist, Merck KGaA, Darmstadt, Germany) was spin-coated on the silicon wafer and patterned with a maskless UV aligner (μPG 501, Heidelberg Instruments Mikrotechnik GmbH, Heidelberg, Germany). The silicon wafer was etched 10 μm deep, and concave electrode patterns were formed. The SEM image of the cliché is presented in Fig. 2(b). For printing, PEDOT:PSS was coated on a PDMS sheet (Fig. 2(c1)), and the layer of PEDOT:PSS was partially transferred to the cliché (Fig. 2(c2)). As the electrode patterns of the cliché are concave, the electrode patterns of the PEDOT:PSS layer remain on the PDMS sheet. Finally, the patterned PEDOT:PSS layer is transferred to the prepared Parylene/EL composite, and the structure in Fig. 2(a3) is obtained. The photograph of the printed electrodes pattern is presented in Fig. 2(d). The fabricated device is presented in Fig. 3.

3. Results

First, by applying and releasing the stretch, wrinkle formation and its stretchability were experimentally verified. Following the method in [15], equi-biaxial stretch was induced in the device. As illustrated in Fig. 4(a), the device was fixed using a ring-shape jig and a plate and stretched by inflating the device. The amplitude of the stretch λ was estimated by measuring vertical deformation of
the device \( h \) (the relationship between \( \lambda \) and \( h \) is available in [15]: \( \lambda = 1.2, 1.4, \) and \( 1.9 \) at \( h = 5, 7, \) and \( 10 \) mm, respectively). The stretch \( \lambda \) was defined as the ratio of the current length and the initial length (With no stretch, \( \lambda \) is 1). Figure 4(b) presents the zoomed image of the surface after applying and releasing the stretch (\( \lambda = 1.4 \)). The wrinkle structure of the Parylene and PEDOT:PSS layers emerged on the surface. As presented in Fig. 4(c), with re-stretching, the wrinkle structure flattened and no apparent fracture was observed on the electrodes.

Figure 5 shows that an AC voltage (approximately, 1 kV at 30 kHz) was applied to the device, and light emission was observed in the condition with and without stretch. For applying a voltage, strips of conductive tape were attached to the surface above the electrodes. Although the protective layer functions as an insulating layer, AC voltage generates an AC electric field between the electrodes. In Figs. 5(a1) (in a normal room) and 5(a2) (in a dark room), the light emission of the device in the condition without stretch is shown. As shown in Figs. 5(b1) (in a normal room) and 5(b2) (in a dark room), with equi-biaxial stretch \( \lambda = 1.4 \), the part between the electrodes emits light. The images show that the luminosity of the device under stretching was reduced in comparison to that without stretch. The reduction under stretch was caused by the increase in the gap between the electrodes, which resulted in a reduction in the amplitude of the electric field. Through the experiment, two-dimensional stretchability of the light-emitting device was verified.

4. Discussion

We previously reported that an increase in the weight ratio of the EL powder to PDMS enhances luminosity [16]. In this study, a weight ratio of 5:1 was used for the stretchability of the device. As the weight ratio of EL powder increased, the PDMS/EL powder composite became more fragile. In addition, the weight ratio of the EL powder is related to the smoothness of the film of the composite. The more EL powder the composite contains, the surface of the film become less smooth, and the PEDOT:PSS layer on it becomes more fragile. The relationship between the smoothness and stretchability will be analyzed in future studies. Concerning the luminosity of the device, the intensity of the electric field depends on the gap between the electrodes. Technically, reverse offset printing enables line and space on the level of single-micrometer [17,18]. Although the breakdown voltage of the Parylene and PEDOT:PSS layers should be considered, the device has room for improvement with respect to its luminosity, which can be done by narrowing the gap of the electrodes and amplifying the intensity of the electric field between them. In this study, the electrodes were patterned in a comb-shape. With the printed electronics technique, electrodes in another shape can also be printed. For example, if line and space patterns are printed on both the top and bottom surfaces of the functionalized elastomer, a light-emitting device with passive matrix electrode can be fabricated.

5. Conclusion

In this study, by utilizing the functionalized elastomer and comb-shape electrodes, stretchable devices were fabricated. The PEDOT:PSS layer maintains its conductivity under equi-biaxial stretch \( \lambda = 1.4 \), and the electric field between the electrodes induces light emission. For fabrication, both Parylene and PEDOT:PSS layers were deposited on non-stretched elastomer, whereas in the previously reported method, thin film layers were deposited on pre-stretched elastomer [13]. Because no stretching was required during the film deposition, functional layers (i.e., conductive layer) were coated and patterned with the printed electronics technique, which potentially enables cheap production over a large area [19,20].

Stretchable light sources are in demand for use in health care and deformable displays. Our study helps the development of methods for fabricating stretchable light sources.
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