CARBON NANOTUBES-ECOFLEX NANOCOMPOSITE FOR STRAIN SENSING WITH ULTRA-HIGH STRETCHABILITY

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ABSTRACT

We developed highly stretchable, flexible and very soft conductors based on the carbon nanotubes (CNTs)-silicone rubber (Ecoflex®) nanocomposite thin films. The resistance of the CNTs-Ecoflex nanocomposite thin film was recovered to its original value under cyclic loading/unloading for strains as large as 510%. Failure strain of the CNTs-Ecoflex nanocomposite was measured to be about ~ 1380% showing its ultra-high stretchability and robustness. As an application of our highly stretchable conductors, we utilized them as skin-mountable and wearable strain sensors for human motion detection. The strain sensors possess high linearity and low hysteresis performance. We observed overshoot behavior of the strain sensors with maximum normalized overshooting peaks 15%. Finally, motion detection of the finger and wrist joints was conducted by using CNTs-Ecoflex nanocomposite thin film strain sensors.

INTRODUCTION

Resistive type strain sensors respond to the mechanical deformation by the change in electrical resistance. Resistive type flexible strain sensors have been reported by coupling of polymers and nanomaterials such as silver nanoparticles (AgNPs) [1], silver nanowires (AgNWs) [2, 3], carbon black particles (CBs) [4, 5], carbon nanotubes (CNTs) [6, 7], and graphene [8, 9]. Moreover, polymeric materials and nanomaterials have been utilized as flexible substrate and strain sensing elements, respectively. For instance, highly stretchable strain sensors have been reported by using AgNWs/PDMS nanocomposite thin films [2, 3], CNTs/PDMS composites [6], CBs/ thermoplastic elastomer (TPE) nanocomposites [4], CBs/silicone rubber (Ecoflex®) composites [10], and graphite/natural rubber composite thin films [7]. However, nonlinear response with high hysteresis is one of the main drawbacks of the reported resistive type sensors [2-4, 6, 7, 10]. On the other hand, highly stretchable (ε>100%) and high performance strain sensors are needed for the wearable and skin-mountable applications such as rehabilitation and personal health monitoring [11-13], sport performance monitoring [14, 15], and entertainment fields (e.g. motion capture for games and animation) [2, 16]. The change from "homogeneous" thin film to the "inhomogeneous" thin film is found to be the reason of high nonlinearity. For example, in the case of the AgNW network/PDMS nanocomposite based strain sensors, we found that homogeneous AgNW network changes to the inhomogeneous network under high strains inducing bottleneck locations in the percolation network [2]. These bottlenecks critically limit the electrical conductivity through percolation network causing a high nonlinear resistance-strain dependency of the strain sensors. Highly non-uniform crack propagation in the thin film is the reason of highly nonlinearity of the graphene woven fabrics (GWFs)/PDMS composite based strain sensors [8]. In addition, high hysteresis behavior of flexible strain sensors could be explained by two mechanisms: (i) intrinsic hysteresis behavior of polymeric materials itself due to their viscoelastic properties, and (ii) interaction between nanomaterials and polymer substrates [2, 6]. Specially, in the case of nanocomposite films, friction force between nanomaterials and polymers plays an important rule on the hysteresis performance. Moreover, under stretching, sliding of nanomaterials occurs within polymer medium. Upon releasing cycle, percolation network of nanomaterials cannot re-establish itself suddenly due to the friction force causing high hysteresis behavior in the strain sensors [2]. Therefore, achieving high performance resistive type strain sensors is still challenging.

Herein, we report highly flexible and stretchable thin films based on the nanocomposite of CNT percolation network and Ecoflex. The CNTs-Ecoflex nanocomposite thin films have been utilized as wearable and skin-mountable strain sensors for human motion detection. The characteristics of strain sensors such as linearity, hysteresis performance, stretchability, and overshoot behavior have been investigated. Strain sensors possess high stretchability (ε ~ 510%) with very good resistance recovery under cyclic loading/unloading. Strain-resistance dependent behavior of the strain sensors is highly linear with low hysteresis. Strain sensors show overshooting upon stretching with maximum overshoot peak value of 15%. As applicability of our strain sensors for the human motion detection, we conducted finger and wrist joint motion detections. The resistance changes caused by the bending and relaxation of strain sensors on the finger and wrist were measured in real-time with high repeatability, stability and fast response.

MATERIALS AND FABRICATION

Multi-walled carbon nanotubes (MWCNTs) with an average length and diameter of 5~20 μm and 16 ± 3.6 nm were purchased from Hyosung Co., South Korea. 0.05 %wt. of non-functionalized CNTs was added into isopropyl alcohol (IPA) and sonicated for an hour. The CNTs-IPA solution was further stirred for another hour to release agglomerated CNTs and perfectly suspend all CNTs into the IPA medium. The uniform suspension of CNTs in IPA was stored for further experiments.

Figure 1 shows fabrication process of the highly stretchable strain sensors based on the CNTs-Ecoflex nanocomposite thin films. CNT solution was first drop-cast on the patterned polyimide (PI) substrate (i.e. rectangular shape: 3×40 mm) and the solution was dried under light heating. Light heating (Lamp light: OSRAM DR 51 50W 12V with Luminous intensity of 1450 cd) was utilized to provide a uniform evaporation of IPA and homogeneous deposition of the CNT thin film [2, 3]. After formation of the CNT thin film, it was annealed under 150 °C for 30 min to remove residual organics. Filtration method was used to transfer the CNT thin film to the polymer medium [2, 17]. To avoid soaking of the liquid Ecoflex between connected CNTs, CNT thin film was pressed by a PI stamp to make it more dens and uniform [17]. Next, liquid Ecoflex with thickness of 0.5 mm was cast on the CNT thin film. Liquid Ecoflex penetrated into the porous network of the CNT thin film due to its very low viscosity. After curing the liquid Ecoflex, a robust nanocomposite of CNTs and Ecoflex was fabricated. All CNT thin film were successfully transferred to the surface of Ecoflex simply by peeling-off the Ecoflex layer from PI substrate due to the strong interfacial adhesion between CNTs and Ecoflex. Then, copper wires were attached to the two ends of the thin film by silver paste and another layer of Ecoflex with the same thickness (~ 0.5 mm) was cured on the top of the CNT thin film to form sandwich structured samples (i.e. Ecoflex layer/CNTs-Ecoflex nanocomposite thin film/Ecoflex layer).

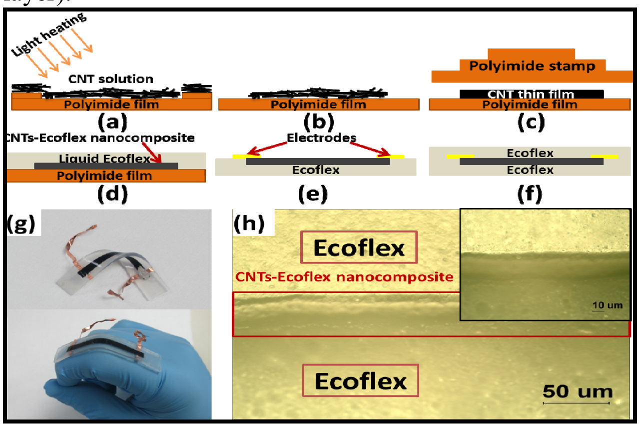

Figure 1: a-f) Fabrication processes of the CNTs-Ecoflex nanocomposite thin film. a) Deposition of CNTs on the patterned PI film under light heating. b) Annealing the thin film at 150 ℃ for 30 min. c) Pressing the thin film by a PI stamp to make it more dense and uniform. d) Casting the liquid Ecoflex on the CNT thin film and curing it at 70 ℃ for 2 hours. e) Peeling-off and flipping the Ecoflex layer and attaching the electrodes by silver paste. f) Casting another layer of Ecoflex and curing it at 70 ℃ for 2 hours. g) Photographs of the fabricated samples under twisting and bending. h) Cross-sectional optical images of the sandwich structured sample; nanocomposite layer is embedded between two layers of Ecoflex.

RESULTS AND DISCUSSION

Figure 1g shows the fabricated strain sensors under twisting and bending with high flexibility and softness. They could easily be attached onto the clothing or directly mounted on the skin. Figure 1h depicts cross-sectional optical image of the sandwich structured sample. As the figure shows, thin layer of the CNTs-Ecoflex nanocomposite is sandwiched between two layers of Ecoflex.

Electromechanical tests were conducted by attaching the strain sensors to a motorized moving stage (Future Science Motion Controller, FS100801A1P1). Cyclic loading/unloading was applied to the strain sensors while their current was measured by a potentiometer (CH Instruments, Electrochemical Workstation, CHI901D). Figure 2 shows the typical curve for the relative change of resistance versus applied strain. Different strain levels (i.e. 30, 60, and 90%) were applied to the strain sensor and response of the strain sensors under both stretching and releasing cycles was measured. As figure illustrates the resistance of sample was recovered very well after releasing it from strain with a good linear response. Moreover, there is a small hysteresis in the response of the strain sensor mainly caused by viscoelastic behavior of Ecoflex.

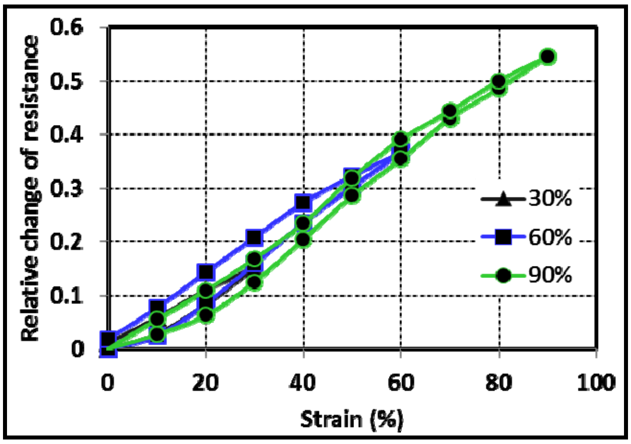

Figure 2: Resistance-strain dependent and hysteresis curve for a CNTs-Ecoflex nanocomposite based strain sensor.

The sensitivity or gauge factor (GF)-slope for curve of the relative change of resistance against applied strain- was calculated to be around ~ 0.61. The main mechanism of resistance-strain dependent behavior of the CNTs/polymers nanocomposite was found to be phenomenon known as “tunneling effect” [18, 19]. Moreover, electrons can tunnel through polymer matrix when the distance between two adjacent CNTs is very small (e.g. 1.8 nm) [18]. When strain sensors are stretched out, re-positioning and re-orientation of CNTs within Ecoflex matrix occur (See inset of Figure 4). Change in the percolation network of CNTs upon stretching increases the tunneling distance between some neighboring CNTs increasing tunneling resistance and consequently increasing the electrical resistance in the whole thin film. Under releasing cycles, recovery and re-establishment of the CNT percolation network leads to recovery of the resistance.

Figure 3 illustrates performance of a strain sensor to the cyclic loading/unloading from 0% to 510%. As shown in figure, there is an excellent overlap between loading profile and response of the strain sensors indicating high stretchability of the CNTs-Ecoflex nanocomposite based strain sensors. The resistance of strain sensors increases linearly with the applied strain and fully recovers to its initial value after releasing with a small hysteresis.

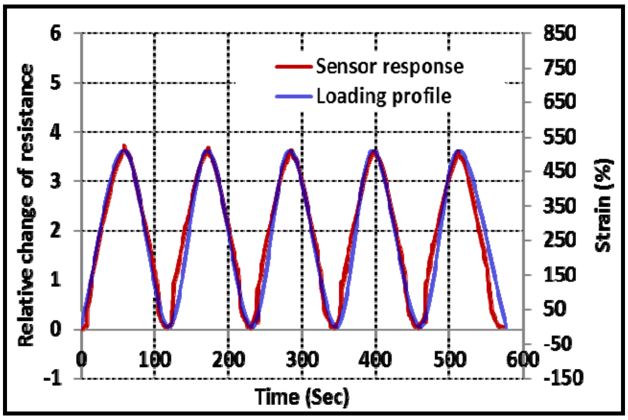

Figure 3: Response of a CNTs-Ecoflex nanocomposite strain sensor to 510% of stretch and release cycles; there is an excellent agreement between sensor's response and loading profile.

To measure the failure strain for a strain sensor, strain was continuously applied to the strain sensor while its resistance changes were monitored. As shown in Figure 4, strain sensor responded to the applied strain with a linear manner and strain sensor lose its electrical conductivity for a strain as large as 1380% indicating ultra-robustness of the CNTs-Ecoflex nanocomposite based strain sensors.

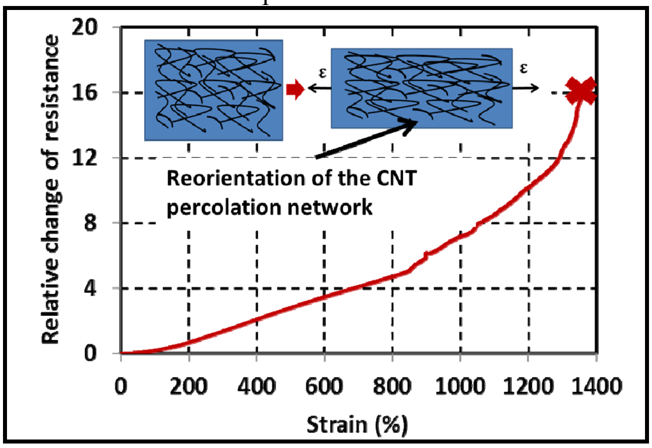

Figure 4: Failure strain measurement for a CNTs-Ecoflex nanocomposite based strain sensor; inset, re-positioning and re-orientation of CNTs in the CNT percolation network upon stretching.

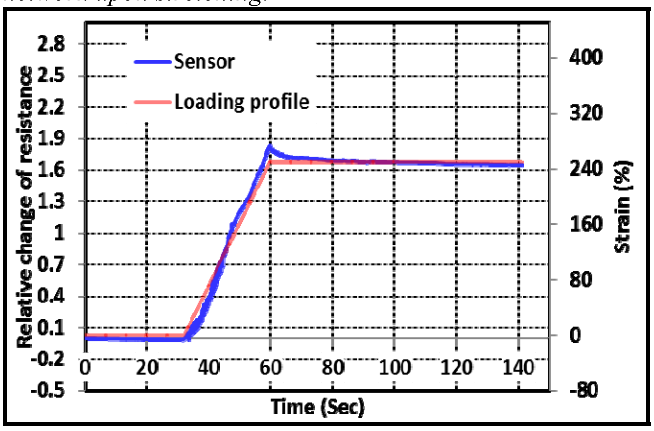

Figure 5: Overshoot behavior of the strain sensor; a small overshoot peak due to the viscoelastic behavior of Ecoflex substrate.

Overshooting was observed in the response of the strain sensors due to the acceleration and viscoelastic properties of the polymer matrix [6, 10]. A strain sensor was subjected to a continuous strain from 0% to 270% and held at strain of 270% while its resistance changes were measured. As indicated in Figure 5, the strain sensor shows overshooting behavior with normalized peak values about 15%.

Since CNTs-Ecoflex nanocomposite based strain sensors are very flexible and stretchable, they could be utilized as wearable and skin-mountable electronic devices. To demonstrate the applicability of our strain sensors, we conducted human motion detection by mounting of strain sensors on the human body. As depicted in the insets of Figure 6, strain sensors are mounted on the finger and wrist for the joint motion detection. Strain sensors are well-attached to the body without any delamination or sliding due to the softness of the Ecoflex matrix. Figure 6 shows the responses of the strain sensors to the bending of finger and wrist. More bending accommodates more strain on the strain sensors and consequently increases the resistance of the strain sensors. Strain sensors responded to the cyclic bending/relaxation with very good repeatability, stability, and fast response.

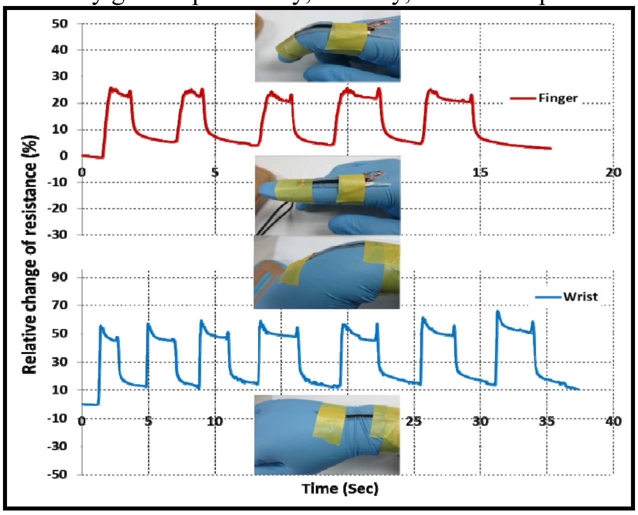

Figure 6: Finger and wrist joints' motion detection using ultra-stretchable and wearable strain sensors; insets of figure, strain sensors mounted on the finger and wrist.

CONCLUSIONS

In this paper, highly stretchable strain sensors based on the CNTs-Ecoflex nanocomposite thin films were fabricated. Stretchability and GFs of the sensors are 510% and 0.61, respectively. The strain sensors exhibit a good linearity with small hysteresis behavior. Maximum normalized overshoot peak was 15% when strain sensors were stretched. As a wearable application, strain sensors were mounted on the finger and wrist for the joint bending measurements. Strain sensors possess high repeatability, stability and fast response under cyclic loading/unloading.

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