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Graphene welded carbon nanotube crossbars for biaxial strain sensors

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A B S T R A C T

Aligned carbon nanotube (CNT) arrays are promising candidates for strain sensors owing to their scalable preparation and excellent conductivity and stretchability. However, aligned CNT arrays are limited by low strain sensitivity and buckling deformation. In addition, cross-stacked CNT array films layer-by-layer assembled on soft substrates exhibit anisotropic mechanical behavior due to their asymmetric layered structures. In this work, we introduced a chemically hybridized CNT-graphene (G/CNT) film in which CNT crossbars are effectively welded together by graphene. The hybrid films demonstrate enhanced isotropic mechanical properties and strain sensitivity with a gauge factor of ~3, together with a high stretchability of more than 20%. The enhanced electromechanical properties are attributed to the improved load transfer efficiency among CNTs by graphene hybridization, as confirmed by Finite Element Analysis (FEA). Biaxial strain sensors based on the hybridized G/CNT films have been applied for sensitive detection of both minute vibrations caused by sound waves and large deformations from finger bending. The sensors were further integrated into a tactile sensing array to map the spatial distribution of the surface pressures.

1. Introduction

Flexible and stretchable strain sensors hold significant promise for applications in wearable and implantable electronics [1–10]. Conventional strain sensors based on metal foils or crystalline silicon are limited by low stretchability [11] (maximum strain of 5%). Recently, there have been numerous interests to develop strain sensors by using nanomaterials and their assemblies. In particular, strain sensors based on random CNT networks respond to tensile strains through intertube rotation and sliding [12–15], which leads to large stretchability. Due to their small tube diameter and very low bending stiffness, the CNTs bend and buckle under compressive strains, which results in irreversible deformation and electrical response to cyclic strains [13,16–18]. Reinforcing the strength of the CNT joints can effectively increase their resistance to buckling deformation [18]. However, at the beginning of the tensile deformation, CNTs in a random network tend to rotate and align along the stretching direction [15,19], which results in minimal changes in their conducting pathways and consequently low sensitivity to small strains [20]. As a result, the gauge factors of strain sensors based on both random CNT networks and reinforced random CNT networks are typically below 0.5 [13,15,17,18].

The microstructures of carbon nanotube assemblies play a critical role in defining their collective properties. Compared to random CNT networks, aligned CNT arrays have demonstrated enhanced electrical and thermal conductivity in the parallel direction owing to the alignment effect [20–22]. In addition, continuous, meter-long aligned CNT array films can be readily drawn from as-grown CNT forests [23]. The facile and scalable preparation of aligned CNT array films, combined with their high electrical conductivity [24,25], is essential for their practical applications in strain sensors [26]. In addition, stretchable, cross-stacked CNT films with isotropic

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electrical conductivity have been prepared through the layer-by-layer assembly of aligned CNT arrays in orthogonal orientation [27]. However, these cross-stacked CNT films exhibit anisotropic electromechanical behavior after transferred onto soft substrates, as a result of their asymmetric layered structures and inefficient load transfer in the out-of-plane direction. It is still a challenging task to design CNT-based strain sensors with isotropic sensing properties, as well as high sensitivity and reliability.

In the present study, cross-stacked CNT films have been chemically hybridized with graphene through chemical vapor deposition (CVD), which effectively improves the load transfer efficiency between the two CNT layers. As a result, the hybrid films demonstrate monotonic electromechanical response along both aligned directions, together with a high stretchability of more than 20%. Furthermore, the strain sensitivity of the cross-stacked CNT films has been increased by 5–10 times after graphene hybridization. Multi-functional biaxial strain sensors have been constructed with the G/CNT hybrid films for sensitive detection of both minute vibrations caused by sound waves and also large deformations from finger bending. A sensor array was further integrated for the spatial mapping of the surface pressure.

2. Experimental

2.1. The synthesis of cross-stacked G/CNT films

The film of super-aligned CNT arrays was drawn from vertically aligned CNT forests [23, 28]. The metallic CNTs are multi-walled with diameter of ca. 10 nm. The cross-stacked CNT films were prepared by sequentially stacking two layers of CNT arrays, with the alignment direction perpendicular to each other, which was then scooped up by a Cu foil. The CNT covered Cu foil was loaded into the quartz tube which was then put into in a horizontal Lindberg/Blue furnace (Thermal scientific). The system was then evacuated to 8.0^*10^{-2} Torr and heated to 1050 °C under a H_2 flow of 8 sccm. After an annealing time of 30 min, a CH_4 flow of 10 sccm was introduced for the growth of graphene. After the graphene growth, the system was cooled down to room temperature with the gases on [29,30].

2.2. The preparation of cross-stacked G/CNT film based strain sensor

The base and curing agent of Polydimethylsiloxane (PDMS, Dow Corning Sylgard 184) were mixed in 10:1 mass ratio. After degassing, the wet PDMS was poured onto a cross-stacked G/CNT film covered Cu and cured at room temperature overnight. The thickness of the PDMS substrate was 2 mm for tensile test and multichannel touch sensing, and was 1 mm for the sound detection test. The Cu foil was then etched with 0.3 mol/L $FeCl_3$ aqueous solution. The membranes were rinsed twice with distilled water to remove $FeCl_3$ residual, and then dried in air. Silver wires were attached to two ends of the cross-stacked G/CNT film for electrical measurements.

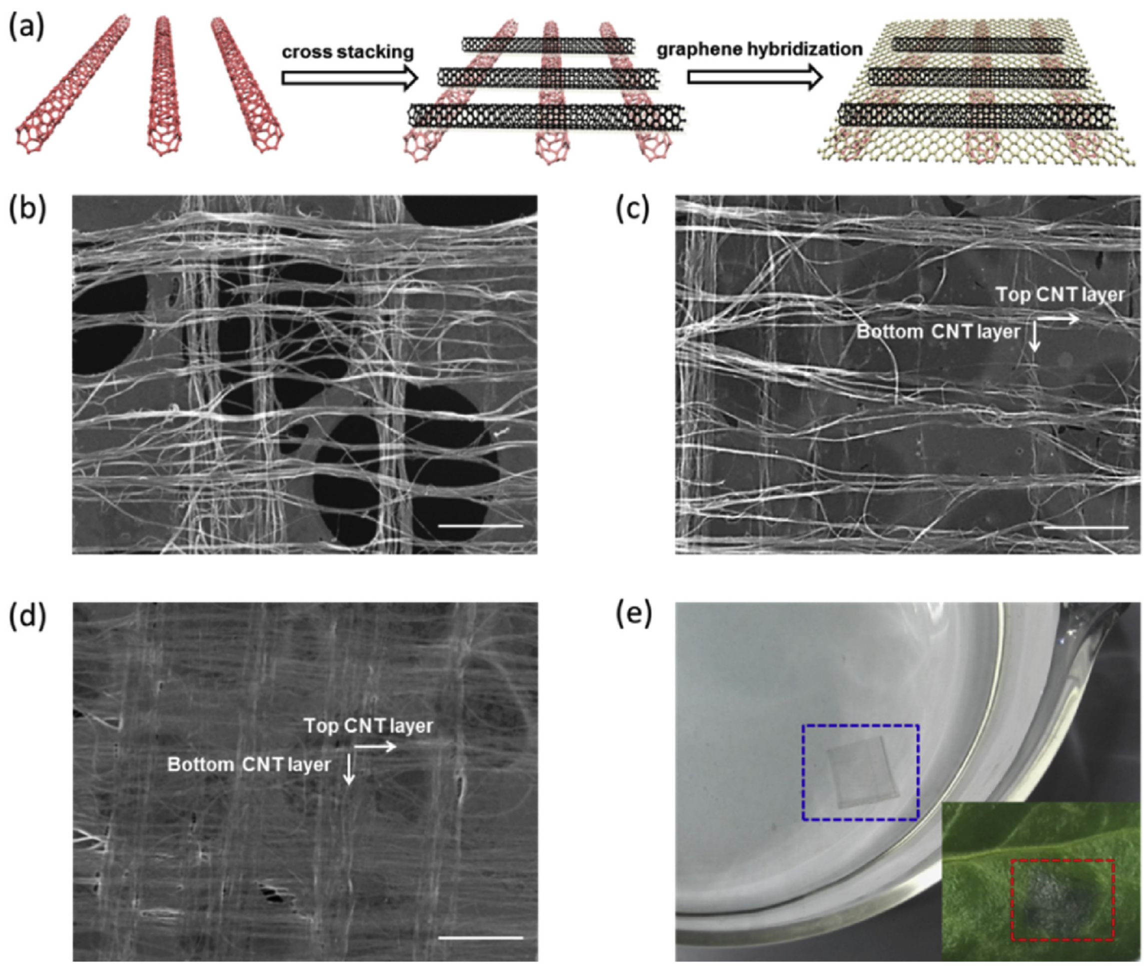

Fig. 1. The preparation of cross-stacked G/CNT hybrids. (a) The schematic of the preparation process. (b) SEM image of a cross-stacked CNT film on Cu grid. (c,d) The SEM images of cross-stacked G/CNT hybrid film on Cu grid from the view of the top and bottom. The alignment direction of different CNT layers is marked by arrows. Scale bar: 2 μm. (e) The optical image showing a freestanding cross-stacked G/CNT film on water. Inset: a cross-stacked G/CNT film conformally contacted onto the rough surface of a leaf. (A colour version of this figure can be viewed online.)