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Fabrication of well-controlled wavy metal interconnect structures on stress-free elastomeric substrates

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A B S T R A C T

We propose a new technique for fabricating well-controlled wavy surface structures on an elastomeric substrate at a few-micrometer scale without any pre-stretching and deposition steps, as the platform for stretchable metal interconnects. In this process, the wavy structure is defined by photolithography on a stress-free elastomeric substrate, so that we can provide various types of wavy profiles for metal interconnects with arbitrary sizes and orientations within a single substrate. As the wavy structures can be formed only within selected regions while keeping the whole substrate area free of strain, it may be possible to fabricate entire circuitry including active devices directly on the elastomeric substrate with no need for mechanical transfer steps. The present technique can provide a practical strategy for realizing large-area stretchable electronic circuits, for various applications such as stretchable display or wearable electronic systems.

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1. Introduction

Stretchable electronics has been considered as a promising technology for overcoming the limitations of conventional rigid electronic systems, in various applications such as foldable or stretchable displays [\[1,2\]](#page--1-0), implantable or wearable biodevices [\[3,4\],](#page--1-0) as well as sensor skins for robots [\[5\]](#page--1-0). As a strategy for realizing highly stretchable electronic circuits that maintain proper functions under a large amount of tensile strain, the interconnects have been either produced as metal lines having stretchable configurations [\[6–9\]](#page--1-0), or made from conductive elastomeric composites [\[10,11\]](#page--1-0). Especially, among many approaches for producing stretchable metal interconnects on elastomeric substrates, the employment of wrinkled wavy structures has been widely investigated [\[12–14\]](#page--1-0).

In previous methods for fabricating wrinkled metal interconnects [\[12–14\]](#page--1-0), a supporting elastomeric substrate is first stretched, then metal interconnects are deposited or transferred onto the prestrained substrate, and finally the prestrain is released, resulting in spontaneous formation of wrinkled wavy electrodes. When an external stretching force is applied, the strain can be accommodated by relaxation of the prestrain, accompanied by flattening of wrinkled structures. Although such prestrain-based techniques have been demonstrated to be very successful in achieving high stretchability [\[12–14\]](#page--1-0), they become less practical as the specimen area is increased because it is difficult to precisely control the structure of wavy profiles over a very large substrate area [\[15\].](#page--1-0) The non-uniform distribution of prestrain also makes it difficult to obtain high reliability, especially in the vicinity of pattern or specimen edges [\[15\]](#page--1-0). Moreover, as it is nearly impossible to perform device fabrication processes (such as photolithography, thin film deposition or etching) directly on a strained substrate, the circuit devices have to be pre-fabricated on a mother substrate and transferred all at once, which should be quite difficult for large area or high density circuits [\[12–14\]](#page--1-0).

In the present work, we propose a new process for fabricating well-controlled wavy surface structures on an elastomeric substrate at a few-micrometer scale, as the platform for stretchable metal interconnects. In this process, the wavy structure is defined by photolithography, so that we can provide various types of wavy profiles for metal interconnects with arbitrary sizes and orientations. In addition, as the wavy structures can be formed only within selected regions while keeping the whole substrate free of strain, it may be possible to fabricate entire circuitry including active devices directly on the elastomeric substrate with no need for mechanical transfer steps. The present technique can provide a practical strategy for realizing large-area stretchable electronic circuits, for applications as stretchable display or wearable electronic systems.

2. Experimental procedures

[Fig. 1](#page-1-0) shows the fabrication process of the elastomeric substrate with a wavy surface profile and stretchable metal electrodes. First,

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Fig. 1. Fabrication process of a wavy metal interconnect. (a) Patterning of alternating photoresist lines and spaces. (b) Anisotropic plasma etching of Si with $\mathrm{SF}_6/\mathrm{O}_2$ gasses. (c) Formation of wavy surface profiles by coating of a thin photoresist layer. (d) Casting of a PDMS layer on the wavy mold. (e) Production of a wavy PDMS substrate. (f) Deposition of the metal electrode layer on the wavy PDMS substrate. (g) Coating of a PDMS capping layer on the metal electrode.

patterns of alternating photoresist lines and spaces were formed on a Si substrate by conventional photolithography, where the line-width and space were identical to each other $(\sim 5 \,\mu m)$ (Fig. 1a). By anisotropic plasma etching of Si with $SF₆/O₂$ gasses, periodic trench structures with square-like cross-sections were produced (Fig. 1b). After removing the photoresist etch mask, another photoresist layer was spin-coated on the whole substrate, for smoothening trench edges and producing a wavy mold structure (Fig. 1c). Then, by casting a 10:1 mixture (weight ratio) of polydimethylsiloxane (PDMS) prepolymer and curing agent (Sylgard 184 Silicone Elastomer Kit, Dow Corning) onto the mold and curing at 60 \degree C for 2 h in an oven, the wavy surface profile was replicated on the elastomeric PDMS layer (Fig. 1d). By releasing the PDMS layer from the mold, a stretchable substrate with a wavy surface profile was obtained (Fig. 1e). For forming stretchable metal electrodes, Ti (=50 Å) and Au (=500 Å) films were sequentially deposited on the wavy PDMS substrate through a shadow mask by electron beam evaporation (Fig. 1f). The electrodes had a 'dogbone' shape with two 7×7 mm² pad regions connected by a 2mm-wide and 30-mm-long line segment. During the metal deposition step, all types of electrode samples (with flat and wavy surface profiles) were loaded together, so that metal films on each sample can be deposited simultaneously in a single deposition process. Then, a capping PDMS layer was additionally coated on the metal electrode layer except contact pad areas (Fig. 1g). For the stretching test, the electrode sample sheet cut into a 20×60 mm² size was loaded onto a house-made stretching stage, where the two pad regions were fixed on the stage and electrically connected to a Keithely 4200 parameter analyzer by copper wires. We stretched the electrode manually by a step of 0.1 mm at a rate of 0.1 mm/s, holding at each position for 2 s for measuring the electrical resistance. All parameters including the sample dimension, initial position, and deformation rate were carefully controlled, so that there was no difference among the test conditions for each type of electrode samples.

3. Results and discussion

[Fig. 2](#page--1-0) show the scanning electron microscopy (SEM) images of photoresist/Si molds with wavy surface profiles. Although evenly-spaced Si trenches have square-like cross-sections after dry etching by SF_6 and O_2 gasses, their corner and edges become rounded by the photoresist layer additionally coated, leading to sinusoidal surface profiles. In [Fig. 2a](#page--1-0) and b, the mold has a onedimensional (1D) wavy profile propagating only along the x-direction, while an 'egg-plate' structure with two-dimensional (2D) waves could also be produced as shown in [Fig. 2c](#page--1-0) and d. The period, amplitude or direction of the wavy profiles can be adjusted at a few-micrometer scale by varying those of Si trenches, which can be achieved simply by controlling the Si pattern size or dry etching conditions.

By replicating the wavy structures of the mold onto the PDMS layer, a stretchable elastomeric substrate having wavy surface profiles was successfully produced without any pre-stretching or metal deposition steps, as demonstrated in [Fig. 3](#page--1-0). Furthermore, as the surface structure was defined by conventional photolithography, we could also form locally flat regions within the wavy-structured area ([Fig. 3c](#page--1-0) and d). In applications of the wavy elastomeric substrate as a platform for stretchable electronic circuits, those flat islands can provide proper sites for active devices. In previous schemes using the pre-stretching and transfer processes [\[12–14\],](#page--1-0) transferred active devices are inherently exposed to a certain amount of compressive stress after the pre-strain is released, which limits the amount of pre-strain or causes the degradation of circuit performances [\[14\].](#page--1-0) By locating actives devices on the stress-free flat regions while the external force is mainly compensated by wavy metal interconnects, a higher stretchability can be achieved. Furthermore, if the active device area is selectively stiffened by controlling the local cross-linking density [\[16\]](#page--1-0) or mechanical reinforcement [\[17\],](#page--1-0) the reliability of stretched circuits would be improved remarkably. In this case, the whole circuitry can be fabricated directly on the elastomeric substrate without employing the transfer processes.

In a previous work, Xiao et al. fabricated a wavy Si mold for PDMS, first by forming a periodic sawtooth structure on Si using anisotropic KOH etching, and then converting it into a sinusoidal shape by spin coating a thin photoresist layer [\[18\].](#page--1-0) Using this technique, they successfully produced a PDMS substrate with wavy profiles at a few or tens of micrometer scale, as a substrate for stretchable Au electrodes. In their method, however, as the initial sawtooth structure is formed by anisotropic Si etching that occurs in accordance with the crystallographic orientation, it is nearly impossible to produce wavy profiles with arbitrary directions within a single substrate or control the shape, amplitude and wavelength independently. On the other hand, Jeong et al. fabricated a wavy aluminum substrate by a machining process, and used it as a mold for forming 1D-wavy PDMS substrates [\[19\]](#page--1-0). They also demonstrated that Ag electrodes with the wavy profile (of hundreds micrometer scale) provide good stretching performance. However, due to some intrinsic limitations of the machining process employed for the mold fabrication, it is difficult to produce more complex wave patterns or reduce the pattern size further.

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