# Auxetic Superelastic TiNiCuCo Sputtered Thin-Films For Stretchable Electronics

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## Introduction

Auxetic shape memory alloy (SMA) materials offer many opportunities for ballistics<sup>1</sup>, deployable antennas<sup>2</sup>, actuators, stretchable electronics<sup>3-5</sup>, and biomedical devices<sup>6,7</sup>. Auxetic materials are periodic structures characterized by a negative Poisson's ratio, meaning they expand laterally when stretched longitudinally. Structures that have a negative Poisson's ratio are known to exhibit many seminal material properties including synclastic bending, high stiffness-to-weight ratio, high impact strength, reduced fatigue crack propagation, and vibration damping<sup>8</sup>. These properties make auxetic structures attractive for stretchable electronic sapplications that are intended to interface with 3D biological surfaces. Such wearable and implantable electronic systems are desired to have a large area density (e.g. regions for functional device integration) and must withstand large levels of applied external strain (~30 % - 100 %) without compromising the functional components of the device<sup>6</sup>. The major drawback of traditional auxetic structures is that the expandability and compressibility of the entire structure are limited by 1) the stiffness of the connecting hinges of the structures and 2) the mechanical properties of the material. The stiffness of traditional auxetic structures may limit



Fig. 1 Mechanical behavior of novel "rotating serpentine rectangle" and "rotating archimedean rectangle" auxetic geometries. A) Demonstration of auxetic behavior of the structures under compression. B) Structures at equilibrium. C) Demonstration of auxetic behavior while structures are stretched to large macroscopic tensile axial strains. (Unpublished Research)

their practical use as implantable medical devices, or for other wearable bioelectronic applications, as the stretchable requirements cannot be met with traditional designs.

One fabrication technique used in stretchable electronics is the "island interconnect", which involves structuring a series of periodic islands connected at the center by stretchable interconnects. The stresses on the islands remain low even when deformed because the stresses and strains are concentrated in the stretchable connections<sup>9</sup>. Examples of such interconnects include serpentines<sup>4,10</sup>, fractal patterns, and archimedean spiral structures<sup>9</sup>. However, there are also some limitations to traditional stretchable designs, including a positive (or zero) Poisson's ratio. In addition, it is usually necessary for traditional stretchable electronics to be integrated on an elastomeric substrate to promote elastic recovery.

The work presented here will show the mechanical properties of auxetic structures are significantly improved by using sputtered superelastic shape memory alloy materials compared to traditional materials (e.g. Cu, Si). In particular, sputtered freestanding TiNiCuCo SMAs offer many advantages as substrates for stretchable electronics<sup>4,5</sup>. Previously this SMA was demonstrated to be ultra-low fatigue able to transform reversibly to a superelastic strain of 2.5% for up to 2 x  $10^7$  cycles<sup>11,12</sup>. Two novel auxetic structures with enhanced expandability and compressibility are presented, fabricated from sputtered TiNiCuCo. The novel geometries presented in this work are based on the combination of the auxetic rotating rectangle structure<sup>8</sup> where the rotating hinges are replaced by two common stretchable interconnects (e.g. serpentines and archimedean spirals). The influence of functional fatigue on the electrical properties, thermal-induced and stress-induced phase transformations of the novel stretchable auxetic TiNiCuCo thin-films will be presented.

#### **Experimental Results and Discussion**

Lima de Miranda<sup>13</sup> developed a freestanding SMA fabrication process in 2013 that allows for the development of freestanding SMA films with thickness between 10  $\mu$ m – 80  $\mu$ m, and allows the patterning of complex microstructures with feature sizes as small as 10  $\mu$ m. This process was used to fabricate novel stretchable auxetic structures with TiNiCuCo, shown at equilibrium in Fig 1B. These auxetic structures are called "rotating serpentine rectangle" and "rotating Archimedean rectangle"<sup>6</sup>, herein referred to as the "serpentine" auxetic structure and "archimedean" auxetic structure for short. The dimensions of the free islands in both geometries in Fig. 1 are 1.0 mm x 1.25 mm, and both structures had a TiNiCuCo film thickness of 53  $\mu$ m. The archimedean spiral had a wavelength ( $\lambda$ ) of 1.25 mm, peak-to-peak amplitude (A) = 2.2 mm, and width (w) = 50  $\mu$ m, and the serpentine interconnect had similar dimensions ( $\lambda = 1 \text{ mm}$ , A = 1 mm, and w = 50  $\mu$ m). Fig. 1A) shows the maximum negative Poisson's ratio of -0.77 and -0.18 is obtained after applying uniaxial compressive strains of -19.5 % and -47% to the serpentine and archimedean structures, respectively. Poisson's ratio is calculated by measuring the change in the transverse strain divided by the change in axial strain in the structure. Fig. 1C) shows the serpentine and archimedean structures still retain auxetic behavior under tension, with a Poisson's ratio of -0.17 and -0.16, when stretched to external strains of 23 % and 90 %, respectively. These values of axial strain are quite large considering an "S-shaped" auxetic structure with reduced stress concentrations could only maintain auxetic behavior when stretched to external strains of 15 %<sup>14</sup>. Similar to the



Fig. 2 A) Novel rotating archimedean rectangle auxetic geometry showing enhanced stretchability. The archimedean spiral displayed superlelastic behavior when tested under both compression and tension conditions between -50 % to 200 % axial strain for 100 cycles. B) Rotating archimedean rectangle auxetic geometry under 265 % tensile axial strain. C) Demonstration of superelastic recovery of TiNiCuCo, with 3.75 % plastic strain due to remnant stress induced martensite. (*Unpublished Research*)

"S-shaped" auxetic structures, both the serpentine and archimedean structures demonstrated a change from negative Poisson's ratio to a positive (or zero) Poisson's ratio when stretched to larger tensile axial strains than the values given in Fig. 1C).

The superelastic response of TiNiCuCo fabricated into the novel archimedean auxetic geometry is shown in Fig 2A). Tensile tests reveal TiNiCuCo could reversibly stretch between -50 % to 200 % macroscopic axial strain for 100 cycles with no fatigue. The serpentine structure was also able to superelastically stretch between -30 % to 50 % for 100 tested cycles with no fatigue (unpublished research, data will be shown). All stress concentrates in the stretchable interconnects, with little to no force on the islands. Extremely low forces are obtained for the archimedean spiral auxetic geometry compared to the serpentine geometry. This means there is a relatively small amount of SMA material transforming along the

spirals. This makes the archimedean spiral geometry attractive for stretchable applications requiring large stretchability levels. While the archimedean spiral can deform significantly more than the serpentine, it comes at the cost of out-of-plane deformation. Fig. 2B) shows the archimedean structure at a maximum tested axial strain of 265 %. At this axial strain, the maximum macroscopic elongation in the archimedean spirals is 454 %. After removal of the applied load, Fig 2C) shows superelastic recovery of the structure with a relatively low amount of plastic strain (e.g. plastic axial strain of 3.75 %). According to FEM simulation results, copper thin-films with the same dimensions as the novel archimedean auxetic structures can only elastically stretch by an axial strain of 15 % (data will be shown), while TiNiCuCo was experimentally shown to superelastically recover at least 200 % axial strain<sup>6</sup>.

### Conclusions

The novel stretchable auxetic geometries "rotating archimedean rectangle" and "rotating serpentine rectangle" structures fabricated from superelastic TiNiCuCo can be used as substrates for stretchable electronics. Both structures are proven to be simultaneously highly stretchable and retain auxetic behavior under large levels of external tensile and compressive strains. The stress concentrates in the stretchable interconnect, while the islands remain under low stresses. This means the islands spaces are available to further device integration. The MEMS compatible fabrication process developed by Lima de Miranda et al. could allow future monolithic integration of functional MEMS

components onto the islands of the superelastic stretchable auxetic substrates. The use of superelastic SMA materials as the substrate might eliminate the need to integrate stretchable MEMS devices onto elastomer substrates in the future.

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