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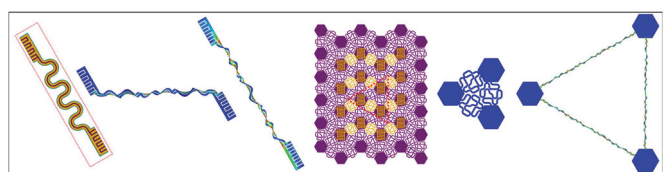
Mechanical designs employing buckling physics for reversible and omnidirectional stretchability in microsupercapacitor arrays

Renxiao Xu, Yonghui Zhang and Kyriakos Komvopoulos 

Department of Mechanical Engineering, University of California, Berkeley, CA, USA

ABSTRACT

Stretchable electronics draw widespread attention with reported applications in various sectors, including health care, optoelectronics, and energy. However, irreversible interconnect deformation and direction-dependent stretchability may greatly limit the longevity and functionality of many stretchable systems operating under multidirectional, repetitive loading and unloading conditions. In this work, we introduce mechanical designs that can significantly enhance reversible, omnidirectional stretchability in a typical microsupercapacitor array. Simulation results from a series of computational studies demonstrate that structural buckling followed by out-of-plane deformation of interconnects are the fundamental physical mechanisms responsible for the increased stretchability. The present analytical methodology provides a computational framework for the effective design of other electronic systems with demanding deformability requirements.



IMPACT STATEMENT

This manuscript contains original work on stretchable electronic devices designed to sustain reversible and omnidirectional excessive stretching, bending, and twisting of supercapacitor arrays by the physics of structural buckling.

ARTICLE HISTORY

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KEYWORDS

Finite element analysis; microsupercapacitor arrays; stretchable electronic materials; omnidirectional stretchability; reversible deformation

Introduction

Stretchable electronic systems have drawn widespread research attention and commercialization in recent years, mainly as a result of advances in materials discovery and mechanical design [1,2]. For many of these systems, functional elements can maintain full electronic performance due to the combination of a well-designed component layout and soft, highly deformable interconnects and encapsulations. Because of their high-performance stability under large deformation, stretchable electronics have found applications in many fields, including epidermal health monitoring [3–6], *in vivo* stimulation and surgery [7,8], bioinspired electronic skins [1,9,10], and biomimetic imaging [11]. For all these systems, developing a power supply module with compatible stretchable mechanics is of paramount importance. In previous

studies, stretchable solar cells [12–14], batteries [14–16], and supercapacitors [17,18] have been investigated as candidates for power supply modules. Compared to batteries, supercapacitors demonstrate higher power density, greater safety during operation, and longer device lifespan.

In a recent study [17], it was demonstrated that a supercapacitor array (see the image in Figure 1(a)) can be uniaxially stretched by 150% while maintaining full electrochemical performance. This result was achieved by joining rectangular supercapacitor units with arc-shaped interconnects and encapsulating the entire circuit layer (i.e. supercapacitors and interconnects) in honeycomb-shaped polydimethylsiloxane (PDMS) substrates and superstrates. Similar to other stretchable systems with ‘island-bridge’ design, the supercapacitor units

(islands) remain almost undeformed, while interconnects (bridges) provide all the stretchability through in-plane bending when the entire module is under uniaxial tension. However, yielding of the metal interconnects rendered the deformation of this system irreversible. According to that study, the maximum principal strain ($\sim 8.2\%$ for 150% uniaxial elongation) in metal (Au) interconnects was much higher than the yield strain ($\sim 0.3\%$), although lower than the fracture strain of Au ($\sim 10\%$). Because of this irreversible deformation, the life-cycle of the stretchable supercapacitor array is expected to be much shorter than what is desirable for stretchable electronics (~ 1000 cycles). Based on a commonly used criterion in finite element analysis validated by cyclic mechanical testing and four-probe resistance measurements [19–22], i.e. the maximum principal strain exceeding the yield strain in half of the width of an interconnect section, it was found that the reversible uniaxial stretchability of this supercapacitor array is only 8%. Another disadvantage in this supercapacitor array design is that both the reversible uniaxial stretchability

ε_r (i.e. the elongation at the yield strain of interconnect) and the total uniaxial stretchability ε_t (i.e. the elongation corresponding to a 5% maximum principal strain in interconnect) are significantly higher in one direction than others. As a result, the supercapacitor array is compliant and stretchable in the x -direction ($\varepsilon_r = 8\%$ and $\varepsilon_t = 150\%$), but stiff and much less deformable in the y -direction ($\varepsilon_r = 0.19\%$ and $\varepsilon_t = 3.1\%$). These two major drawbacks, i.e. irreversible interconnect deformation and direction-dependent stretchability, may significantly limit the application of the supercapacitor array in stretchable electronics, where operation requires significant multidirectional cyclic loading and unloading.

The objective of this study was to identify alternative, improved design strategies of this typical supercapacitor array by employing the physics of structural buckling. It is shown that one design, referred to as *reversibly stretchable* – design A (Figure 1(b)), provides at least 54 times higher reversible uniaxial stretchability and also greatly improved total uniaxial stretchability than what was previously reported [17], whereas another design, referred

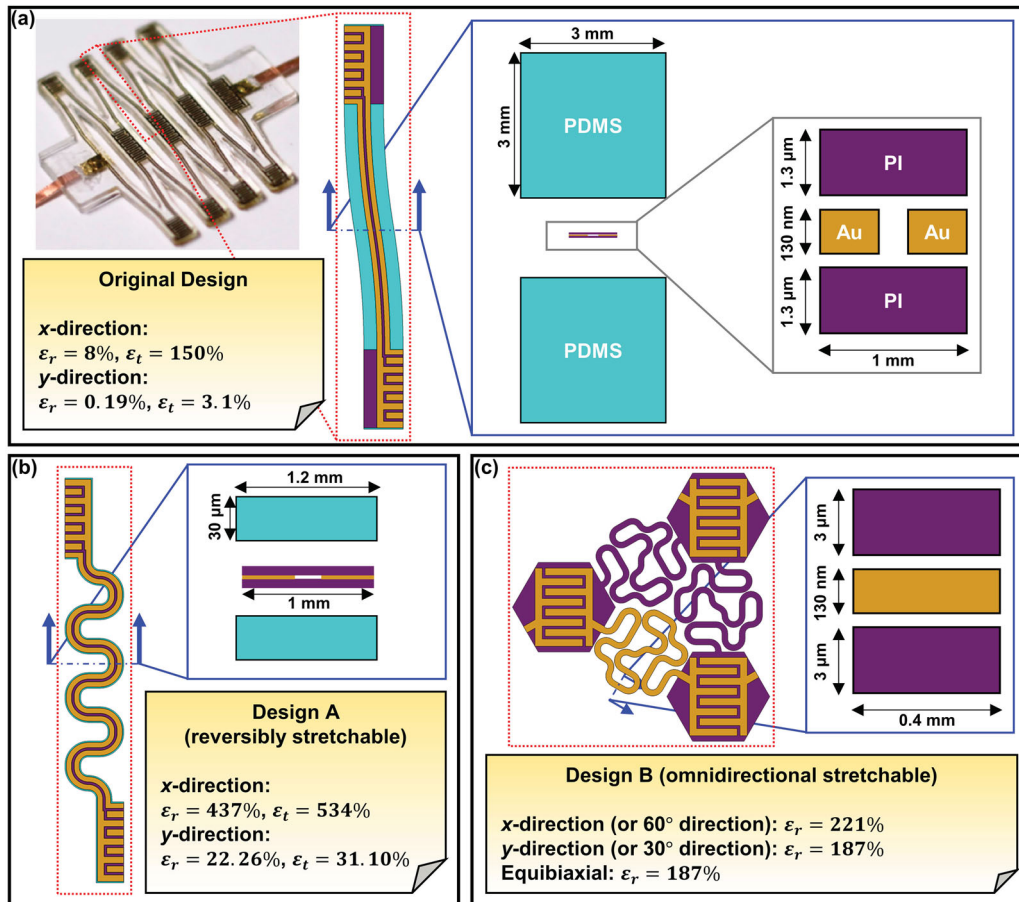


Figure 1. Schematics of three stretchable supercapacitor designs shown by their constituent unit cells. (a) Original design including a PI/Au/PI composite layered structure encapsulated in PDMS [17]. (b) Improved design A. (c) Improved design B. In each panel, all unit cells are shown in top view (red dash boxes) and side view (blue and gray solid boxes). Side views are not drawn to scale. ε_r and ε_t denote reversible stretchability and total stretchability, respectively.

to as *omnidirectional stretchable* – design B (Figure 1(c)), offers high reversible uniaxial stretchability ($\geq 187\%$) in all four planar directions examined, as well as high reversible equibiaxial stretchability (187%). Both of the proposed designs are supported by finite element analysis (FEA) results.

Results and discussion

Previous studies have shown that buckling and post-buckling deformation of thin films can greatly improve the reversible stretchability of interconnects [15,19,20]. Because the critical buckling strain (which is proportional to the thickness squared) of such thin interconnects is small, out-of-plane deformation commencing upon buckling significantly reduces the strain energy in metal interconnects. As a result, interconnect yielding occurs at much larger stretchability. However, for interconnects in the stretchable supercapacitor array of Pu et al. [17] (a representative periodic unit cell of that supercapacitor array is shown at the top of Figure 1(a)) buckling does not occur even after interconnect yielding because the top and bottom PDMS encapsulation layers are too thick. Therefore, to reduce the critical buckling strain, it is necessary to significantly reduce the beam thickness. This can be accomplished by using thinner PDMS encapsulation layers. The blue curves in Figure 2(a) and (b) show the critical buckling strain ε_c versus beam thickness t for identical planar dimensions of interconnects (including both the polyimide (PI)/Au/PI composite and the PDMS encapsulation) with those of the device shown in Figure 1(a). It can be seen that ε_c decreases sharply from $\sim 25\%$ to $\sim 0.7\%$ in the x -direction (Figure 2(a)) and from $\sim 3\%$ to $\sim 0.006\%$ in the y -direction (Figure 2(b)), respectively, as the beam thickness decreases from 600 to 60 μm . In addition to the beam thickness, ε_c is also affected by the in-plane geometry. To further reduce ε_c , an alternative design A is proposed (Figure 1(b)), where the curved beam formed by two joined arcs is replaced by a narrower (the interconnect width is reduced from 3 to 1.2 mm) and much longer serpentine beam with multiple turnings. For this design, ε_c (red curves in Figure 2(a) and (b)) is consistently much lower than the original design for all beam thicknesses examined due to the increased structural compliance of the serpentine beam. Notably, for $t = 60 \mu\text{m}$, beam buckling in design A commences at an elongation in the x -direction of $\sim 0.032\%$ or an elongation in the y -direction of $\sim 0.002\%$.

After buckling, interconnects accommodate further uniaxial elongation by post-buckling deformation, until reversible stretchability is terminated by the onset of yielding. Figure 2(c) and (d) show the reversible stretch-

ability ε_r and total stretchability ε_t of unit cells versus beam thickness t for the original and serpentine planar designs in the x - and y -direction, respectively. The reversible stretchability of the unit cell with a serpentine beam design increases from 80% to 437% in the x -direction and from 0.6% to 22.26% in the y -direction as the beam thickness decreases from 500 to 60 μm . Similarly, an increase in reversible stretchability from 51% to 270% in the x -direction is achieved by the unit cell with curved beam patterns when the beam thickness is reduced from 400 to 60 μm ; however, the reversible stretchability in the y -direction remains low and constant.

The monotonic dependence of reversible stretchability on beam thickness can be explained by the nature of post-buckling deformation. For the two unit cells examined, post-buckling deformation comprises the superimposed effects of rotation, translation, bending, and twisting of each segment of the deformed interconnects. These out-of-plane deformation routes are energetically favorable because they result in minimum strain energy. For thinner beams, the strain energy due to bending and twisting are both lower, so that out-of-plane deformation dominates the overall deformation of the structure. Since small strain energy also implies small strain in the metal interconnect, the onset of plastic yielding is deferred to a larger deformation. Therefore, a larger reversible stretchability is obtained with a unit cell having thinner interconnects. It is noted that the reversible stretchability of unit cells with thick beams (e.g. 600 μm) is significantly lower than that of beams with smaller thickness. In addition, the reversible stretchability in the y -direction of unit cells with the original planar pattern remains very low and constant ($\sim 0.19\%$) despite the significant decrease in beam thickness. The reason is that these beams (open symbols in Figure 2(c) and (d)) do not exhibit out-of-plane deformation with increasing elongation because the strain energy is higher than that due to in-plane deformation; thus, the beams of the original design undergo only in-plane deformation and the interconnects yield prematurely.

A comparison of unit cells with original and serpentine designs reveals that the latter design yields higher reversible stretchability at all beam thicknesses. This is because the reduced width of the serpentine beam provides more space for accommodating complex turnings, resulting in interconnects with a much longer initial length. These long interconnects composed of more deformable segments significantly enhance the capacity for large reversible stretchability. The total stretchability ε_t of unit cells in the x - and y -direction versus beam thicknesses t was also obtained for both planar pattern designs (Figure 2(c) and (d)). Similar

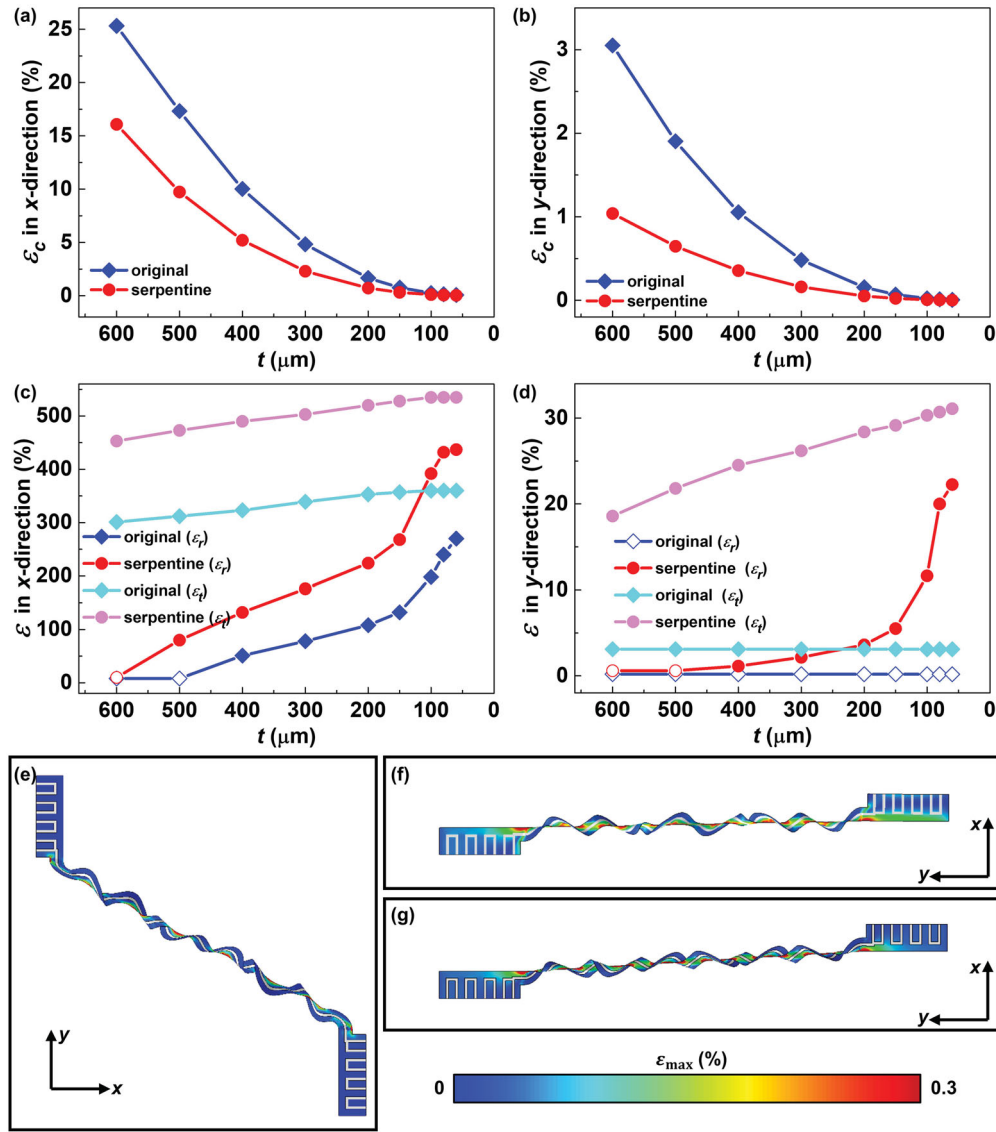


Figure 2. Buckling and post-buckling deformation of interconnects with original and serpentine interconnect patterns. (a) Critical buckling strain ε_c in the x -direction versus beam thickness t for interconnects with original (blue curve) and serpentine (red curve) patterns. (b) Critical buckling strain ε_c in the y -direction versus beam thickness t for interconnects with original (blue curve) and serpentine (red curve) patterns. (c) Reversible stretchability ε_r and total stretchability ε_t in the x -direction versus beam thickness t for interconnects with original and serpentine patterns. (d) Reversible stretchability ε_r and total stretchability ε_t in the y -direction versus beam thicknesses t for interconnects with original and serpentine patterns. (e–g) Deformation of structure with improved design A elongated (e) uniaxially in the x -direction, (f) uniaxially in the y -direction, and (g) equibiaxially up to the reversible stretchability limit. The color scale shows the maximum principal strain in metal interconnect.

to reversible stretchability, the total stretchability also reaches a maximum for unit cells with thin ($t = 60 \mu\text{m}$) serpentine interconnects. Because design A offers much higher reversible stretchability, i.e. $\varepsilon_r = 437\%$ in the x -direction (Figure 2(e)) and 22.26% in the y -direction (Figure 2(f)), and total stretchability, i.e. $\varepsilon_t = 534\%$ in the x -direction and 31.1% in the y -direction, it represents a greatly improved design of reversibly stretchable supercapacitor arrays. Compared to the original design, design A with exactly the same areal coverage and layout of supercapacitor ‘islands’ and the same width of

the metal portion of interconnects offers significantly increased reversible uniaxial stretchability (~ 54.6 times higher in the x -direction and ~ 117 times higher in the y -direction) and higher total uniaxial stretchability (~ 3.6 times higher in the x -direction and ~ 10 times higher in the y -direction).

Although design A provides a significant increase in reversible uniaxial stretchability compared to the original design, its stretchabilities in the x - and y -direction differ by a factor of ~ 19.6 . The fact that design A is much more stretchable in one direction than another is not

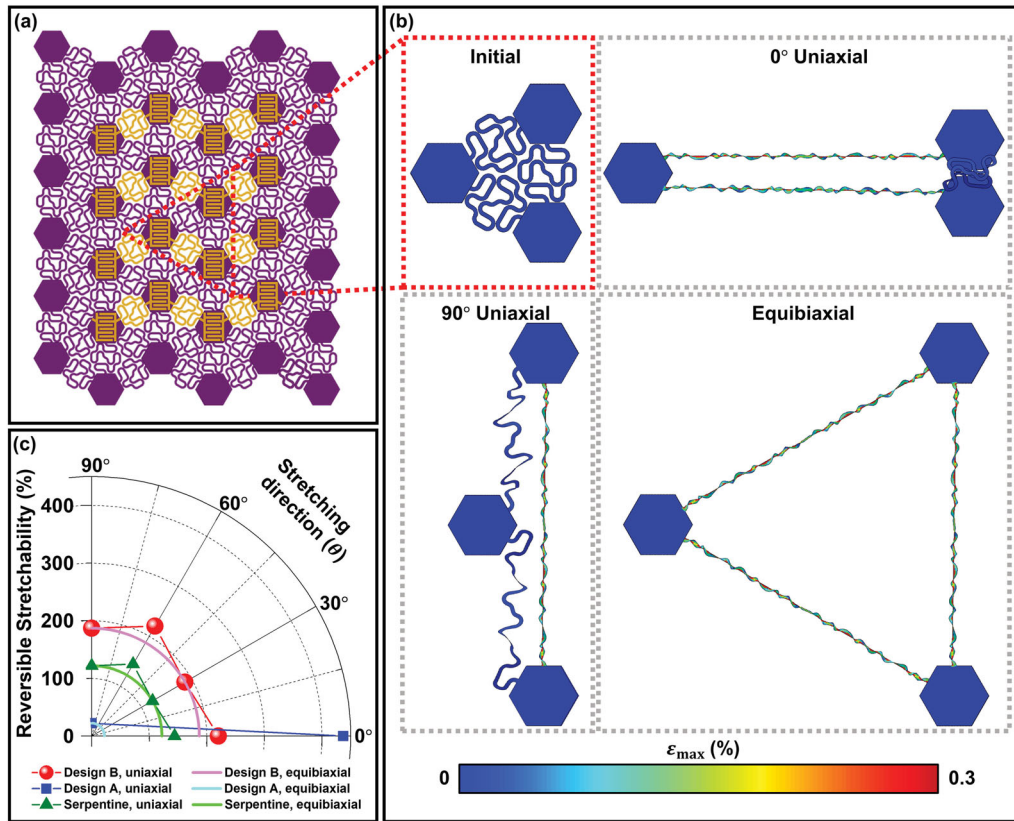


Figure 3. Analysis of improved design B. (a) Top-view schematic of a stretchable supercapacitor array with design B as part of an infinite two-dimensional tessellation. (b) Deformation of a periodic unit cell of design B uniaxially elongated in the 0° (top right) and 90° (bottom left) stretching direction and equibiaxially (bottom right) to the reversible stretchability limit. The color scale provides the maximum principal strain in the metal layer. (c) Reversible uniaxial (points) and equibiaxial (curves) stretchability of design A, design B, and a design with serpentine interconnects versus stretching direction θ .

compatible with applications requiring sufficient uniaxial stretchability in all loading directions. In addition, some applications may require electronic systems to exhibit equibiaxial stretchability, which is usually limited by the stretchability in the least deformable direction. As shown in Figure 2(g), the reversible equibiaxial stretchability of design A is only 22.26% due to the limited stretchability in the y -direction. Therefore, for applications requiring omnidirectional stretchability, i.e. equally stretchable in all planar directions and equibiaxially stretchable, an alternative design must be considered. A feasible design B is shown in Figure 3(a). This design is inspired by the art of two-dimensional tessellation, where a flat plane is tiled by regular hexagons without gaps or overlaps. While previous studies have used the idea of two-dimensional tessellation to develop stretchable bio-inspired triangular/honeycomb/Kagome network composite materials, these designs usually include only deformable filamentary units [23,24]. A stretchable supercapacitor array, however, has both non-deformable supercapacitors ('islands') and deformable interconnects ('bridges') as building blocks. In design B, one-fourth of the hexagons

serve as 'islands' for supercapacitors to reside, where each rectangular supercapacitor is inscribed in one hexagonal island. Other hexagons (i.e. surrounding 'islands') are filled with serpentine-shaped interconnects ('bridges'). These long, compliant, and thin interconnects serve both as mechanical joints for islands and as electrical connections for supercapacitors and can accommodate elongations in different directions through buckling followed by out-of-plane deformation. The interconnects for electrical connections are composite beams consisting of a 130-nm-thick Au layer sandwiched between two 3- μm -thick PI layers (Figures 1(c) and 3(a)), whereas interconnects used as mechanical connections are 6- μm -thick PI beams (shown in purple in Figure 3(a)). As shown in Figure 3(c), the reversible uniaxial stretchability for design B in four representative in-plane angular directions θ is 221% at $\theta = 0^\circ$ (x -direction) and 60° and 187% at $\theta = 30^\circ$ and 90° (y -direction). The deformation details of a representative unit cell are shown in Figure 3(b). FEA analysis also shows that design B exhibits 187% reversible equibiaxial stretchability. Because of the relative high ($\geq 187\%$) and fairly uniform (only $\sim 17\%$ difference) reversible

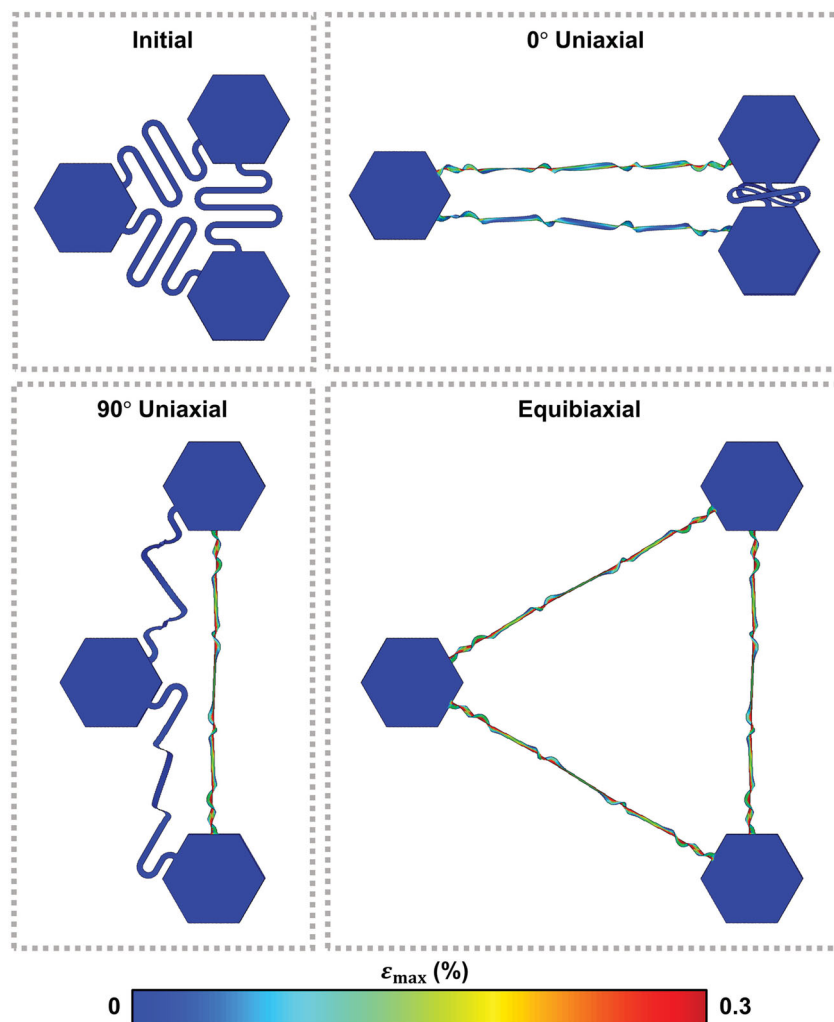


Figure 4. Deformation of a periodic unit cell of a design with serpentine interconnects uniaxially elongated in the 0° (top right) and 90° (bottom left) stretching direction and equibiaxially (bottom right) to the reversible stretchability limit. The color scale shows the maximum principal strain in the metal layer.

uniaxial stretchability in representative directions and also high (187%) reversible equibiaxial stretchability, design B provides desirable omnidirectional stretchability. Since the areal coverage by supercapacitors in design B (16.4%) is very close to that of the original design and design A (15.5%), while the geometric details of supercapacitors and the widths of metal portion of interconnects are the same, the reversible stretchability of these designs can be compared directly and fairly.

As shown in Figure 3(c) and previously discussed, although design A offers significantly improved stretchability compared to the original design, it is characterized by a nonuniform deformation in different directions. Alternatively, the structure in design B is about equally stretchable in all four planar directions examined and can maintain high stretchability under equibiaxial loading. These features of design B make it suitable for applications requiring high omnidirectional stretchability. It is

also noted that both the tessellation-inspired layout of ‘islands’ and ‘bridges’ and rational choice of interconnect patterns contribute to omnidirectional stretchability. If the interconnect pattern in design B were to be replaced by a classical serpentine pattern with the same interconnect widths (Figures 3(c) and 4), the reversible stretchabilities would be considerably reduced to 144% for $\theta = 0^\circ$ and 60° and 122% for $\theta = 30^\circ$ and 90° .

Admittedly, design B still has some limitations that must be overcome before application in future stretchable electronics. For example, the small spacings ($\sim 50 \mu\text{m}$) among the complex interconnect patterns may result in lower yield in photolithography-based microfabrication. Therefore, a more strictly controlled, costly, but still feasible fabrication process is required to prevent self-entanglement of interconnect segments due to residual stress. In addition, the PI clads alone may not provide good electrical insulation and mechanical protection for

the electronics. Soft encapsulation materials with large resistance and high dielectric strength, such as viscous silicone oil [5], ultralow modulus elastomer/adhesives [4,14], or even newer options should be incorporated to ensure reliable packaging.

Conclusions

Improved mechanical designs of a previously reported stretchable supercapacitor array were obtained by allowing for buckling and post-buckling out-of-plane deformation of interconnects. Supported by FEA models, the two proposed designs offer significantly improved stretchable mechanics, although with slightly different targets – one design provides much higher stretchability in the x -direction (design A), while the other design yields much higher omnidirectional reversible stretchability (design B). The results of this study pave the way toward the development of stretchable microsupercapacitors with desirable deformation mechanics. The reported computational procedure may also be applied to the design of other soft (compliant) and deformable electronic systems.

Disclosure statement

No potential conflict of interest was reported by the authors.

ORCID

Kyriakos Komvopoulos  <http://orcid.org/0000-0001-9418-1567>

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