Flexible and Stretchable Devices from Unconventional 3D Structural Design

Hangbo Zhao and Mengdi Han

Northwestern University, Center for Bio-Integrated Electronics, Evanston, IL, USA

Many biological systems in nature are three-dimensional (3D), such as plant roots, animal cells, human vascular networks, etc. The 3D feature provides essential functions in various activities including physicochemical reactions and interactions with the environment. Biomimetic recreation and engineering of 3D architectures inspired by nature has been a topic of increasing interest in recent decades, especially in the area of flexible and stretchable devices. 3D structures on such devices enable new and more complex functionalities as compared to 1D or 2D counterparts. Moreover, the flexibility and stretchability of the substrate provide a unique means of forming, controlling, and tuning 3D structures on it. The interplay between mechanics and structures provides plenty of space and fascinating opportunities for the design and fabrication of unconventional 3D structures with novel functionalities.

In this chapter, various approaches to forming unconventional 3D structures are first introduced, from basic buckling of ribbons, membranes, and non-coplanar bridge-island structures, to more complex deterministic assembly. A series of strategies for extending the level of control in deterministic assembly is discussed, followed by a collection of exemplary flexible and stretchable electronic and optical devices fabricated using 3D deterministic assembly methods.

10.1 Stretchable 3D Ribbon and Membrane Structures Formed by Basic Buckling

Mechanical buckling of thin films under compressive stress provides a powerful route to forming 3D structures. The stretchability of elastomeric substrates is utilized as a platform for buckling micro-/nanostructures. In this section, the fabrication of 3D ribbon and membrane structures based on simple substrate buckling is introduced.
10.1.1 3D Nanoribbons

The John A. Rogers group from the University of Illinois Urbana-Champaign exploited compressive buckling to convert nanoribbons into 3D, wavy structures. The process starts with fabricating patterned thin, flat, single-crystal silicon ribbons on a mother wafer [1]. Then, the Si elements are bound to a pre-strained elastomeric substrate. After peeling off the elastomer and releasing the pre-strain, the silicon is transformed into well-controlled, periodic wavy ribbons that are stretchable. Large-area, uniform-array, wavy nanoribbons can be fabricated using this method. The nanoribbons are highly stretchable, with a maximum strain over 10% for both compression and stretching. A single-crystal silicon p–n diode is showcased as an example of applications of such wavy nanoribbons.

The same principle of buckling nanoribbons could be applied with spatial control over adhesion sites added to create well-controlled local displacement of the ribbons [2]. The fabrication process is schematically illustrated in Figure 10.1a.

Figure 10.1 Buckling of semiconductor nanoribbons for stretchable electronics. (a) Schematic illustration of the fabrication process of 3D buckled nanoribbons. (b) Schematic of buckled nanoribbons in response to stretching and compression. (c) SEM image of GaAs nanoribbons formed. (d) Schematic and optical images of a stretchable photodetector (PD) from nanoribbons. (e) I–V characteristics of fabricated PDs under different stretching degrees. Source: Reproduced with permission from Sun et al. [2]. Copyright 2006, Springer Nature.
An ultraviolet ozone (UVO) mask is used to pattern the surface chemistry on a poly(dimethylsiloxane) (PDMS) substrate, where bonding between the Si or GaAs nanoribbons and PDMS substrate only occurs in lithographically defined regions. The buckled nanoribbons could be embedded in PDMS and undergo extremely high levels of stretching (up to ∼100%), compression (up to ∼25%), and bending (with curvature radius down to ∼5 mm), as shown in Figure 10.1b.

Such stretchable ribbons could be used in the construction of metal–semiconductor–metal photodetectors, as shown in Figure 10.1c–e. The I–V characteristics of the photodetectors are mechanically tunable due to the change of the projected area of the buckled GaAs ribbons.

10.1.2 3D Nanomembranes

In addition to nanoribbons, nanomembranes could also be engineered into 3D configurations using buckling. As shown in Figure 10.2, a silicon thin film, initially on a mother wafer, is transferred to a biaxially pre-strained PDMS slab. After the release of the strain of PDMS, wavy Si nanomembranes are formed with controllable surface topologies.

The formed wavy Si nanomembranes are stretchable, and yield various morphologies in response to different strains applied at different orientations, as shown in Figure 10.2b. These wavy membranes provide a useful path to biaxially stretchable devices.

10.1.3 3D Bridge-Island Structures

More complex structural configurations are achievable by combining ribbons with island-like 2D structures. Figure 10.3 shows an example of non-coplanar, bridge-island structures [4]. The ultrathin circuit mesh fabricated using standard semiconductor fabrication is bound to pre-stretched PDMS at the positions of the flat islands, via ozone surface treatment. Using this strategy, an array of complementary metal–oxide–semiconductor (CMOS) inverters is fabricated and encapsulated with PDMS (Figure 10.3b). The fabricated device offers extremely high stretchability, with stretching strain up to ∼140% and twisting pitch as tight as 90° in ∼1 cm (Figure 10.3c).

The feature size of the stretchable circuit is comparable to conventional 2D devices, while no significant changes in electrical properties are observed owing to the high stretchability of the PDMS substrate and mechanics-guided structural design.

10.2 Deterministic 3D Assembly

While the basic buckling approaches described in Section 10.1 are effective in transforming 2D patterns into 3D configurations, the accessible structures are limited to wavy ribbons, membranes, and combinations of them. In this section, a more complex 3D assembly approach is introduced, namely, deterministic 3D assembly. Deterministic 3D assembly combines lithographically
Fabricate thin Si membrane film

(i) Contact and transfer to pre-strained PDMS slab

(ii) Peel off the PDMS slab; flip over and release the strain of PDMS

(iii) Stretching strain $\varepsilon_{st} = 0\%$

$\varepsilon_{st} = +1.8\%$

$\varepsilon_{st} = +3.8\%$

$\varepsilon_{st} = 0\%$

Figure 10.2 Biaxially stretchable “wavy” nanomembranes. (a) Schematic illustration of the fabrication process. (b) Optical micrographs of wavy Si nanomembranes under different uniaxial strains at three different orientations. Source: Reproduced with permission from Choi et al. [3]. Copyright 2007, American Chemical Society.
controlled layouts of 2D structures with patterned adhesion sites to the surface of pre-strained elastomer substrates to enable fast and scalable assembly of a broad range of 3D structures.

### 10.2.1 Basic Approach of Deterministic 3D Assembly

The basic approach of deterministic 3D assembly is described in detail in Refs. [5–7]. Briefly, the process starts with patterning 2D patterns (precursors) with lithographically defined bonding sites. Then, the 2D precursors are bound to a pre-stretched elastomer slab. Release of the pre-strain induces spatially dependent twisting and bending deformations, transforming the 2D patterns into 3D structures.
As shown in Figure 10.4, 3D conical helices made from single-crystal silicon are fabricated, which were inaccessible 3D architectures previously. The mechanics and shape transformation process can be analyzed and precisely predicted by a finite element analysis (FEA) scheme, and excellent agreement is found between FEA predictions and experiments. A broad class of geometries are realized, including single and multiple helices, conical spirals, flowers, frameworks, and multilevel configurations. Moreover, this 3D assembly approach applies to various material compositions (inorganic semiconductors, polymers, metals, and heterogeneous combinations) and feature sizes (submicrometer to centimeter), as shown in Figure 10.5.

This deterministic 3D assembly approach provides a powerful route to sophisticated classes of 3D structures and materials. The compatibility with the CMOS fabrication methods, and device-grade materials, creates many opportunities for achieving new 3D-structure-based electronic and other devices.

Depending on the basic 3D assembly method described in the previous sections, a series of strategies and processing techniques have been developed to
Figure 10.5 3D structures fabricated using the deterministic 3D assembly approach with various characteristic dimensions and materials. (a) Starfish-jellyfish-like structures. (b) 3D mesostructures of different materials. Source: Reproduced with permission from Zhang et al. [8]. Copyright 2017, Springer Nature.
extend the level of control in the deterministic 3D assembly process, which has greatly increased the space of attainable 3D structures.

10.2.2 3D Kirigami Structure in Micro-/Nanomembranes

The John A. Rogers group from the University of Illinois Urbana-Champaign introduced the concept of kirigami to the 3D assembly method, where 2D micro-/nanomembranes have strategically designed geometries and patterns of cuts [9]. Figure 10.6 lists several examples showing how the 2D silicon membranes with engineered kirigami cuts at precisely defined locations are transformed into 3D structures with strategically determined panel deformations.

10.2.3 Buckling Control Assisted by Stress and Strain Engineering

Engineering of stress and strain in the 2D precursors could also influence the 3D buckling process. Figure 10.7 shows two such engineering strategies. In Figure 10.7a, residual stress is introduced to the 2D precursor by patterning a

[Figure 10.6: Examples of mechanically driven kirigami for deterministic 3D assembly from 2D micro/nanomembranes. (a) A square cuboid. (b,c) Membranes with first- and second-order cross-cuts. (d,e) Membranes with symmetric and antisymmetric cuts. Scale bars, 200 μm. Source: Reproduced with permission from Zhang et al. [9]. Copyright 2015, PNAS.]
2D SiN$_x$ film onto a Si substrate prior to the 2D precursors being buckled [10]. This residual stress, either tensile or compressive, induces different buckling mechanics and results in distinct 3D structures.

Another strategy involves the engineering of elastomer substrates for spatially nonuniform strain in the substrates for buckling. As Figure 10.7b illustrates, the elastomer substrate, which is nonuniform in thickness via molding, induces spatially varying strain upon release of the pre-strain, thereby creating nonuniform 3D structures [11]. A similar strain-engineered method is also used to fabricate 3D microarchitectures based on growth [12].

10.2.4 Multilayer 3D Structures

Instead of single-layered structures, multilayers of advanced materials could also be produced using releasable, multilayer 2D precursors. The schematic illustration of the process is shown in Figure 10.8a [13]. Multilayer 2D precursors are transferred to the stretched elastomer using a layer-by-layer transfer printing
Figure 10.8. Deterministic assembly of 3D mesostructures from multilayer 2D precursors. (a) Schematic illustration of the fabrication process. (b and c) Examples of resulting 3D multilayer structures. Scale bars, 400 μm. Source: Reproduced with permission from Yan et al. [13]. Copyright 2016, AAAS.
10.2 Deterministic 3D Assembly

Complex 3D topologies such as dense architectures with nested layouts are constructed (Figure 10.8b,c). The multilayer feature also enhances the structural stability and drives the motion of extended features in those structures.

10.2.5 Freestanding 3D Structures

A potential disadvantage of the 3D assembly method discussed so far is that the elastomeric substrates, necessary for the assembly process, could impose engineering constraints to the system as the 3D structures are mechanically tethered to the substrates. Several techniques have been developed to bypass this limitation in order to fabricate freestanding 3D structures, including interfacial photopolymerization and wax-assisted transfer [14].

The process of interfacial photopolymerization is schematically shown in Figure 10.9a. A thin, sacrificial Al₂O₃ layer is deposited between the bonding sites and the SU8 droplets. UV light is then used to cure the SU8 droplets, forming the assembled 3D structures. To release the structures, the wax is dissolved, allowing the structures to be transferred to a target substrate. This process can be used to create freestanding 3D structures.

Figure 10.9 Freestanding 3D structures. (a) Interfacial photopolymerization and (b) wax-assisted transfer printing methods for fabricating freestanding 3D structures assembled by compressive buckling. Scale bars, 500 μm. Source: Reproduced with permission from Yan et al. [14]. Copyright 2017, PNAS.
and the elastomer for subsequent release from the substrate. To mechanically
hold the 3D deformed shape, a photodefined epoxy SU8 is cast and cured
onto the 3D structure, followed by patterned backside exposure. Developing
of the epoxy and removal of the Al₂O₃ layer releases the 3D structures into
freestanding objects, with a thin remaining base layer.

For the second method shown in Figure 10.9b, transfer printing is employed,
assisted by wax encapsulation to transfer the assembled 3D structures onto the
target substrate. The wax holds the shape of the deformed structures and is
dissolved upon transfer of the structures onto the adhesive layer on the target
substrate.

### 10.2.6 Morphable 3D Structures by Multistable Buckling Mechanics

3D structures that can undergo reversible transformations are important in a
broad range of applications such as microelectromechanical systems (MEMSs),
biomedical devices, and microrobotics. A scheme in 3D assembly was developed
recently to fabricate morphable 3D structures relying on sequential release of the
pre-strain in elastomeric substrates [15].

The concept of the scheme and representative SEM images are illustrated
in Figure 10.10a,b, respectively. Starting from a biaxially stretched flat state of

![Figure 10.10](image_url)  
**Figure 10.10** Morphable 3D structures by loading-path-controlled mechanical assembly.
(a) Schematic illustration of the strategy with FEA predictions and experimental SEM images of
formed 3D structures. (b) 2D shapes, FEA predictions, and corresponding experimental images
of morphable 3D structures. Scale bars, 1 mm. Source: Reproduced with permission from Fu
the substrate, the pre-strain could be released in two different loading paths: simultaneous release in both \(x\) and \(y\) directions or sequential release (\(y\) direction first, then \(x\) direction). These two loading paths yield two distinct buckling modes and corresponding 3D structures due to the mechanics involved in the buckling process. The entire process is reversible, and thereby the two shape configurations can be switched between each other continuously and repetitively. Again, such reconfigurable 3D structures are achievable in a broad set of materials, multilayer stacks, and over various length scales.

10.3 Flexible and Stretchable Devices from 3D Assembly

The deterministic 3D assembly method discussed in Section 10.2 provides a versatile route to the fabrication of 3D structures made of a broad range of materials, complex geometric configurations, and over various length scales. Such 3D structures are integrated on elastomeric substrates upon assembly, making them capable of dynamically and reversibly changing their shapes. This feature makes them promising building blocks for flexible and stretchable devices for applications spanning from microelectronic, to optical, optoelectronic, and biomedical applications. In this section, some exemplary flexible and stretchable devices fabricated using the deterministic 3D assembly are introduced.

10.3.1 Electronic Devices and Systems

As the fabrication process of the 2D precursors is compatible with the most sophisticated CMOS process, a variety of classes of electronic components, devices, and systems could be made on the basis of the 3D assembly method. Furthermore, the mechanical tunability of the 3D structures gives rise to tunable electronic properties of such flexible devices [8, 13–19].

Several exemplary electronics devices and systems are shown in Figure 10.11. A 3D toroidal inductor is fabricated and can be mechanically configured into two distinct shapes by partial and then complete release of the pre-strain (Figure 10.11a) [8]. Good agreement between the FEA-predicted and experimentally measured inductance as a function of frequency is observed.

In Figure 10.11b, a pyramidal coil is fabricated as part of a 3D NFC device taking advantage of the multilayer stacking strategy [13]. The 3D NFC device exhibits significantly enhanced \(Q\) factor and improved working angle over conventional 2D counterparts.

Figure 10.11c shows a network of Si-nanomembrane-based nMOS transistors interconnected by bridge structures [16]. Helical coils fabricated using compressive buckling are used as effective electrical interconnects for soft electronics, as shown in Figure 10.11d [17].

10.3.2 Optical and Optoelectronic Devices

Complex 3D structures inspired by origami and kirigami are promising platforms for optical and optoelectronic applications. For example, a mechanically
Figure 10.11 Exemplary flexible electronic devices fabricated using deterministic 3D assembly. (a) Measured and modeled frequency dependence of the inductance and $Q$ factor of a 3D torsional inductor. Source: Reproduced with permission from Zhang et al. [8]. Copyright 2017, Springer Nature. (b) A 3D NFC device with enhanced $Q$ factor over conventional 2D counterparts. Source: Reproduced with permission from Yan et al. [13]. Copyright 2016, AAAS. (c) A 3D-interconnected bridge structure with an array of Si nanomembrane nMOS transistors. Source: Reproduced with permission from Kim et al. [16]. Copyright 2018, American Chemical Society. (d) A flexible device composed of a network of helical coils as electrical interconnects. Source: Reproduced with permission from Jang et al. [17]. Copyright 2018, Springer Nature.

Tunable optical transmission window is demonstrated in Figure 10.12a [9]. As the membrane rotates in response to the strain applied to the elastomer substrate, the amount of normally incident light they block also changes continuously, making the device an optical shutter with well-controlled mechanical tunability.

In Figure 10.12b, a 3D hemispherical structure is made by integrating 2D materials (graphene, MoS$_2$) with a buckled hemispherical 3D structure [20]. The 2D semiconductor/semimetal materials provide photodetection and light-imaging capabilities on the 3D structural platform.

10.3.3 Scaffolds as Interfaces with Biological Systems

3D structures formed by compressive 3D assembly could also serve as electronic cellular scaffolds for the growth, recording the stimulation of neural networks.
Figure 10.12 An exemplary flexible 3D-structure-based device for optical and optoelectronic applications. (a) A mechanically tunable optical shutter. Scale bars, 500 μm. Source: Reproduced with permission from Zhang et al. [9]. Copyright 2015, PNAS. (b) A 3D photodetection and imaging system. Source: Reproduced with permission from Lee et al. [20]. Copyright 2018, Springer Nature.
Figure 10.13 shows an example of a freestanding 3D open-cage structure used as scaffold for engineered dorsal root ganglion (DRG) neural networks [14]. The 3D configuration allows interaction and communication with live cells and tissues in 3D. Experiments demonstrate that such scaffolds facilitate the reorganization of cells into hierarchical cellular constructs. This example shows promising opportunities of 3D structures as cellular and tissue scaffolds in fundamental and applied biological studies.

10.4 Summary

This chapter introduces different approaches for forming 3D structures. Bonding nanomembranes in Si or other inorganic materials to a pre-strained elastomer can yield 3D wavy structures with controllable surface topologies upon release of the pre-strain. On this basis, deterministic 3D assembly introduces spatial control of the adhesion sites to further expand the accessible 3D geometries. The final 3D geometries can be tailored through layouts of the 2D patterns, position of the bonding sites, pre-strains, thickness profiles of 2D patterns, loading path, nonuniform elastomeric substrates, and many others. Applications of such 3D mesostructures cover the field of electronics, optoelectronics, optical devices, biointerfaces, energy harvesters, microrobots, and others.
Abbreviations

CMOS complementary metal–oxide–semiconductor
FEA finite element analysis
MEMS microelectromechanical system
NFC near-field communication
PDMS poly(dimethylsiloxane)
SEM scanning electron microscopy

References


