Chapter 2
Mechanical Self-Assembly vs. Morphogenesis

Xi Chen and Jie Yin

Abstract Morphogenesis, as one of the three fundamental aspects of developmental biology, refers to the biological processes of developing certain shapes, which takes place across many length scales, including the morphologies of a cell, a tissue, an organ, and a system. From the intrinsic yet complicated biological and biochemical perspectives, several mechanisms for plant pattern formation have been suggested, such as positional information theory [1] and reaction–diffusion theory [2]. However, the active role of mechanical forces should not be underemphasized.

In the past few years, a great interest has been sparked in the development of biophysical and mechanical theories to explain the plant pattern formation [3, 4]. Among them, the connection of the morphogenetic processes of some plants with mechanical buckling theory receives a great attraction owing to some similarities. Patterns and shape formation are treated as the generation of specific undulating physical topography. From biophysical viewpoints, during the growth of plants, the morphology transition can be treated as spontaneously approaching the pattern/mode with minimal energy, which is similar to the mechanical instability/bifurcation approach. Among the several possible buckling/wrinkling modes (i.e., undulating patterns or structures), the system will spontaneously choose the pattern with the minimized energy.

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2.1 Morphogenesis and Mechanical Buckling Model

A great interest has been sparked in the connection of the morphogenetic processes of some plants with mechanical buckling theory [3, 5–8]. Generally the related studies can be divided into two categories: one is the investigation on the mechanism of phyllotactic patterns in shoots and flowers; the other one is on the shape of leaves.

On phyllotactic patterns, Green pioneered the biophysical explanation for the patterns commonly observed in plant shoots and flowers [3, 4], and proposed the hypothesis that buckling of the compressed tunica is the governing mechanism for determining the local phyllotactic pattern. Dumais and Steele [6] showed that since the sunflower capitulum is under circumferential compression, buckling may be a plausible explanation for the primordium initiation in the captitulum. Shipman and Newell [5] demonstrated that the local phyllotaxis and the deformation configurations on plant surfaces may have resulted from the energy-minimizing buckling pattern of a compressed shell on an elastic foundation.

Several studies have been conducted on the shape of leaves from mechanical principles. Inspired by the similarity between wrinkled edges of torn plastic sheets and a wavy leaf, Marder et al. [9] suggested that some leaves may form wavy edges through spontaneous buckling and proposed a continuum theory on governing similar wavy edges observed in many leaves and flowers. To explain some wrinkled shapes in leaves, Dervaux and Amar [10] proposed an elasticity theory on the morphogenesis of growing tissues, where the growth stress is incorporated into the generalized Foppl–vonKarman (FvK) theory of thin plates. Koehl et al. [11] studied the ecomorphological differences on the blade shapes of kelp, where many species of macroalgae have wide, thin, and ruffled (undulate) blades in sheltered habitats, while their conspecifics at sites with more exposure to rapidly flowing water have narrow, thick, and flat blades. Their research revealed that the change in shape results from elastic buckling induced by mechanical stress. Recently, Liang and Mahadevan [12] studied the shape of long leaves and showed that the typical morphologies with saddle-like midsurface and rippled edges arise from the elastic relaxation via bending following differential growth in leaves. All these studies showed that the different shapes of leaves may result from the differential in-plane deformation within the leaves, which may lead to the occurrence of local or global wrinkling.

2.2 Mechanical Self-Assembly of Single-Layer Film and Multilayered Film–Substrate

In recent years, self-assembled fabrication of micro/nanopatterns and structures involving thin films and multilayers has become the new cornerstone, where there are numerous methods that enable self-assembly, such as surface-tension-based assembly [13, 14], electroactive polymer actuation [15], electric actuation [16], thermal and shape-memory alloy actuation [17], and stress-driven actuation

[18, 19]. In this dissertation, the studies related with mechanical self-assembly fabrication will be reviewed, especially by taking advantages of instability.

The mechanical self-assembly, i.e., the method of creating patterns or structures through utilizing failure of film, has become a focal point in the self-assembly fabrication. There are basically three kinds of failure in thin films: buckling [20], delamination [21], and cracking [22]. Among them, owing to the relative easiness of controlling and manipulation, the study on buckling of a single film sheet [23–26], bilayer thin films [27–30], and film–substrate systems [20, 31–33] has attracted wide interests and found extensive applications in engineering.

2.2.1 Mechanical Self-Assembly of Single- or Mutlilayer Film

Under mismatched deformation or external guidance, buckling may occur in a single-sheet film or multilayer films and leads to different morphologies. The controllable buckling process can be utilized and tailored to create desired patterns and structures. For a single sheet, there are two methods to trigger the self-assembly of a desired pattern or structure: one is the in-plane inhomogeneous and mismatched deformation at different locations; the other one is through the interaction between elasticity and capillarity, i.e., surface-tension-based assembly. For multilayer system, the driving force for the self-assembly mainly comes from the mismatched deformation between the different layers.

Through the programmed shrinkage at different locations of a thin flat gel sheet, Klein et al. [23] created a variety of both 3D large-scale buckling and multiscale wrinkling structures with nonzero Gauss curvatures as shown in Fig. 2.1. Although the resulting structures are not regular and ordered, it shows the promising of forming 3D structures from the buckling of a single 2D sheet.

Through the self-assembled wrapping of a liquid droplet by a planar sheet, Py et al. [24] developed a capillary origami of thin films into 3D structures as shown in Fig. 2.2. They showed that the final encapsulated 3D shapes can be controlled by tailoring the initial geometry of the flat membrane. Through the balance between interfacial energy and elastic bending energy, they revealed the critical length scale below which encapsulation cannot occur, which suggests a new way of mass production of 3D micro- or nanoscale structures. In a recent study it was demonstrated that the resulting 3D structures may offer a promising way to efficiently harvest solar energy in thin cells using concentrator microarrays [25]. A similar capillary origami study was conducted by Patra et al. [26] at the nanoscale using molecular dynamics simulations. They demonstrated that water nanodroplets may activate and guide the self-folding of planar grapheme nanostructures and lead to the self-assembly of nanoscale sandwiches, capsules, knots, and rings as shown in Fig. 2.3.

The mechanical bending and stretching ability of a 2D planar thin film, coupled with or without capillary driving force, may provide us an efficient self-assembly method to create folded 3D micro/nanostructures and devices, which have potential applications as building blocks of functional nanodevices, with unique mechanical, electrical, or optical properties [34].

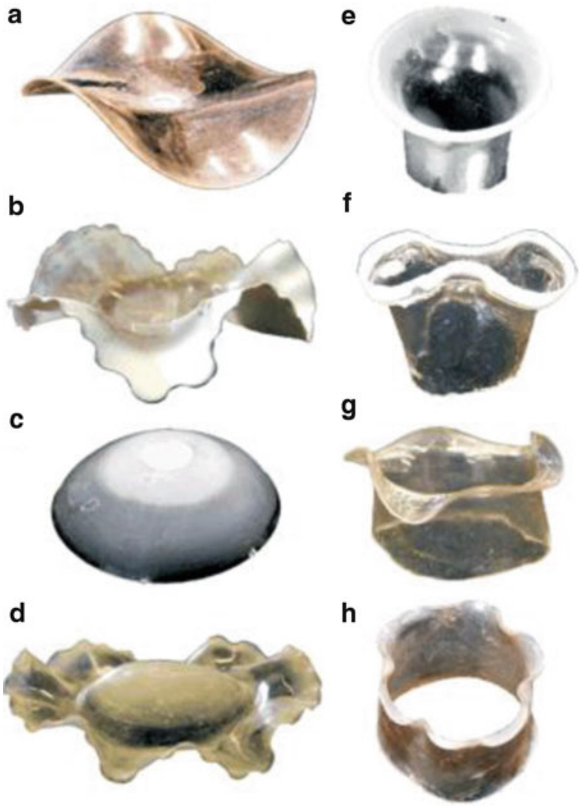

Fig. 2.1 Different structures of sheets with radially symmetric target metrics. Klein et al. [23], reproduced with permission

When combining another different thin film layer with the aforementioned single film sheet, different functional micro/nanostructures can be created through the mismatched deformation between the bilayer system, where such a mismatched deformation can be introduced by misfit lattice strain [29], different rates of thermal expansion [28], and swelling or contraction rate [16].

Schmidt and Eberl [29] pioneered the study on the self-assembly of thin solid films into nanotubes using misfit strain. They showed that when a bilayer of thin films with two different materials is deposited on substrates, after the bilayer is released by selective etching, the bilayer structure would buckle and bend upwards, and finally self-roll into a nanotube driven by the misfit lattice strain between the bilayers, as shown in Fig. 2.4a, b. Through the similar mismatched strain approach, Prinz et al. [35] further created more varieties of 3D micro/nanostructures such as tubes, coils, and helices with width ranging from a few micrometers down to a few nanometers (see Fig. 2.4c).

Despite the promise in creating nanotube or coils, Schmidt and Prinz et al.'s methods are limited to the large misfit lattice strain, which is only applicable to a few materials. By combining the advantages of tailored shape in Py et al.'s study and the self-rolling of tubes in Schmidt's study, recently Gracias and coworkers

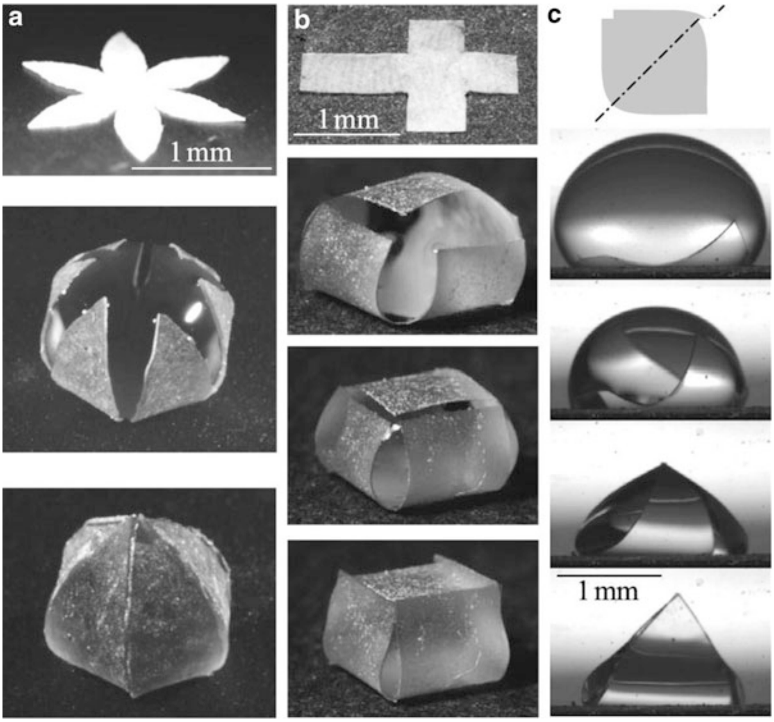

Fig. 2.2 Tuning of the initial flat shape to obtain (**a**) a spherical encapsulation, (**b**) a cubic encapsulation, or (**c**) a triangular mode-2 fold. Py et al. [24], reproduced with permission

[13, 27, 28] proposed and demonstrated a self-assembly method to design and fabricate complex patterned polyhedral micro-containers in the form of cubes, square pyramids, dodecahedra, and octahedral (see Fig. 2.5).

Their strategy uses the thin film sheets as a bilayer hinge consisting of a chromium (Cr)/copper (Cu) bilayer. The self-folding is caused by the residual stress developed during thermal evaporation of the metal thin films, which was due to the mismatch in the coefficient of thermal expansion (CTE) of the bilayer materials. Through the control of the thickness of each layer, the bilayer hinge could fold with certain desired angles and the resulting microstructures have great potential applications as vehicles for drug delivery [36] and 3D electromagnetic components.

2.2.2 Mechanical Self-Assembled Patterns in Film–Substrate System Through Wrinkling

Owing to the constraint of thin film thickness, the self-assembled 3D micro/nanostructures through single or bilayer thin films discussed above prefer the global

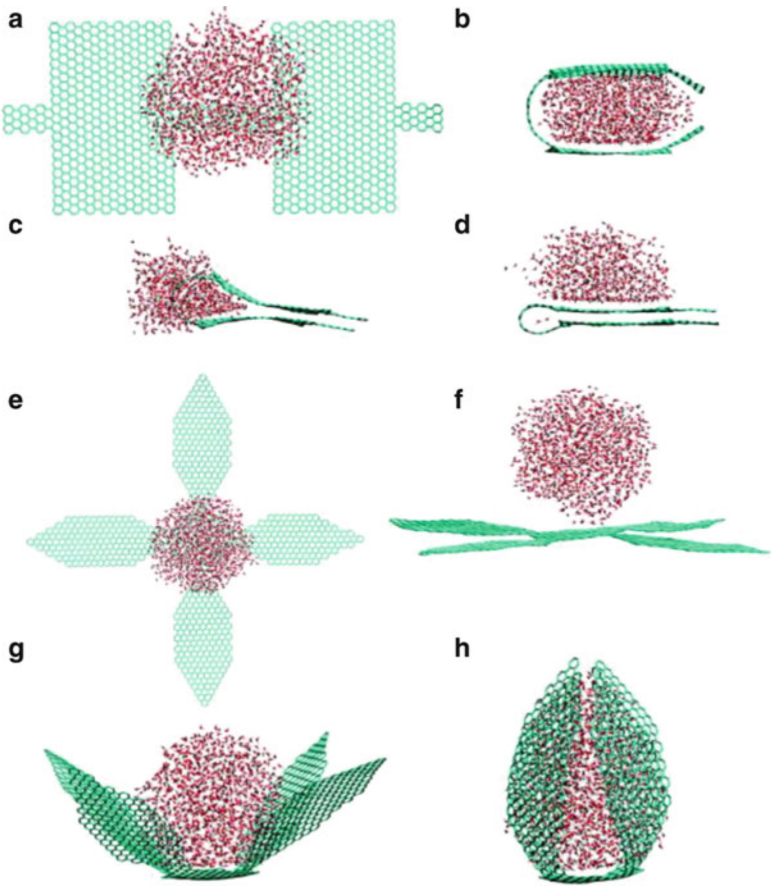

Fig. 2.3 (**a–d**) Water nanodroplet-activated and -guided folding of two graphene flakes connected by a narrow bridge. (**e–h**) Nanodroplet-assisted folding of a star-shaped graphene flake. Patra et al. [26], reproduced with permission

buckling to release the stored strain energy. It favors the global bending deformation with a large wavelength, which is analogous to the Euler buckling of columns. In the bucking of columns, the higher buckling modes with a shorter wavelength are not energetically unfavorable.

However, when the thickness of the underlying substrate is much larger than that of the thin film, since the substrate remains tightly bonded with the thin film during deformation, local wrinkling with a short wavelength will be preferred to release the compressive strain. The resulting wavelength is mainly determined by the competition between the bending energy of the thin film and the stretching energy of the substrates. While the bending energy prohibits the wrinkling with short wavelengths, the substrate favors wrinkles with shorter wavelengths. A trade-off between the bending energy in the film and stretching energy in the

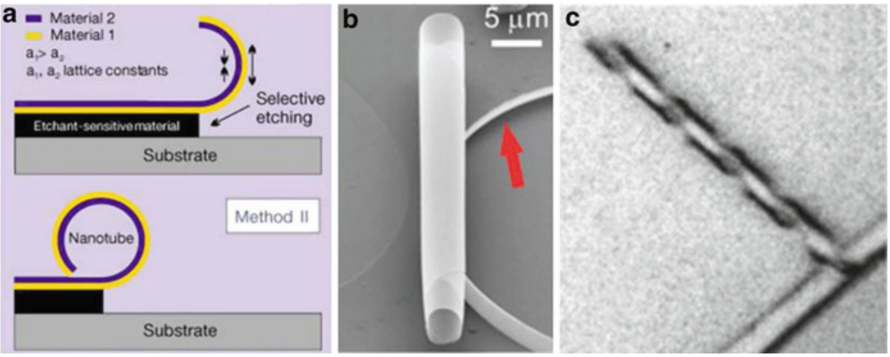

Fig. 2.4 Self-assembly of bilayer thin films (**a**) Schematic illustration of self-rolling of bilayer films under misfit lattice strain. (**b**) The resulting self-assembled nanotube. (**c**) Helix scrolled from a strip. Prinz et al. [35], reproduced with permission

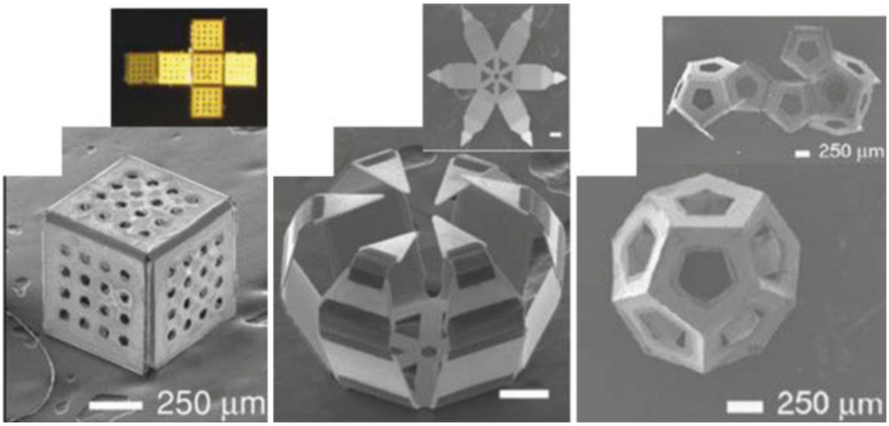

Fig. 2.5 Self-assembled patterned micro-containers through folding of bilayer hinges. On the right corner shows the original planar shape. Reprinted with permission from Bassik et al. [27]. Copyright 2009, American Institute of Physics

substrate determines the optimum wavelength of the wrinkles observed in film/substrate system.

Local wrinkling has been historically considered as a mechanism for structure failure, which should be avoided in the design of sandwich panels [37] and constructions widely used in aerospace and marine engineering [38] as well as the deposition of thin film in the semiconductor industry [39]. In 1969, Allen [37] first studied the problem on the wrinkling of sandwich panels in airplanes with a stiff face rested on a compliant substrate. The usefulness of spontaneous buckling of thin film/substrate systems was first demonstrated by Bowden et al. [20] in 1998, where in their pioneered work they utilized the wrinkling of metal films on soft polydimethylsiloxane (PDMS) substrates to generate ordered micropatterns and

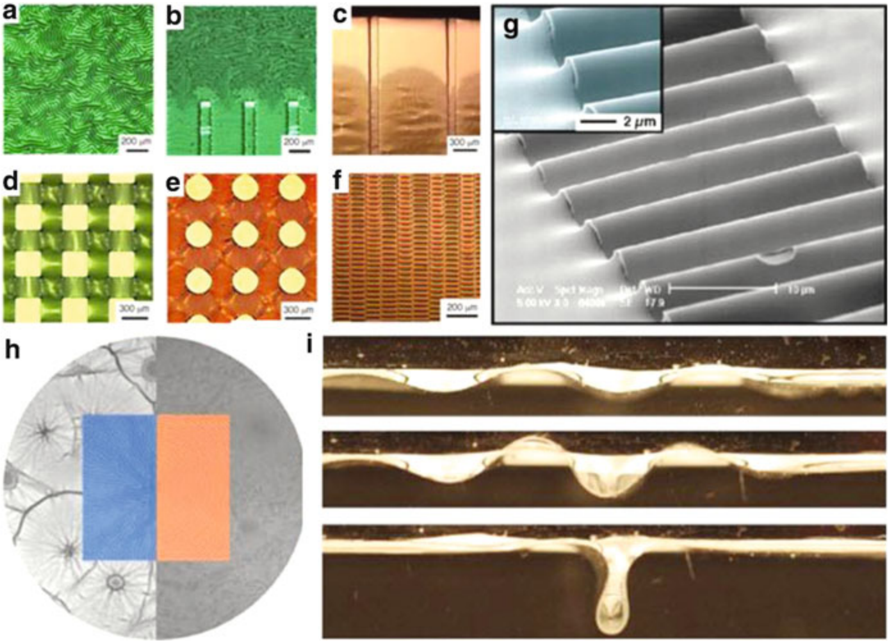

Fig. 2.6 Various wrinkling patterns observed in experiments. (**a**–**f**) Representative patterns in metal films on PDMS substrates through the control of edges and steps, where labyrinth patterns (**a**) far away any steps or edges transit to ordered ones through the rectangular, square, and circular steps or elevations. (**g**) One-dimensional ridged nanoribbons. (**h**) Concentric and radial wrinkled patterns through defects. (**i**) Wrinkle-to-fold transition of thin film on water. Moon et al. [51], reproduced with permission

structures. Spontaneous elastic buckling patterns were observed in the 50 nm-thick gold film as the system was cooled, owing to the mismatched thermal deformation and the films buckled into distinctive patterns, with typical wavelengths on the order of microns. These highly ordered patterns (e.g., Fig. 2.7a) can be precisely manipulated and have found vast applications in stretchable electronics [40–42], MEMS and NEMS [43], tunable optical gratings [44], thin film modulus measurement [45], force spectroscopy in cells [46], control of smart adhesion [47], adjustment of superhydrophobic properties of film [48], and pattern formation for micro/nanofabrication [20, 49] among others.

Following Bowden et al.'s pioneering experiment, extensive experimental, theoretical, and numerical studies were carried out to explore the buckling mechanisms and investigate the feasibility of quantitative control of the patterns for applications in micro- or nanostructures. Among experimental efforts, the substrate surface topology may be manipulated to change the local film stress, so as to generate a variety of ordered patterns (see Fig. 2.6a–f) [49].

Similarly, local physical properties of the thin film can be perturbed to result in various buckle patterns [50], and more refined nanoscale patterns may be achieved

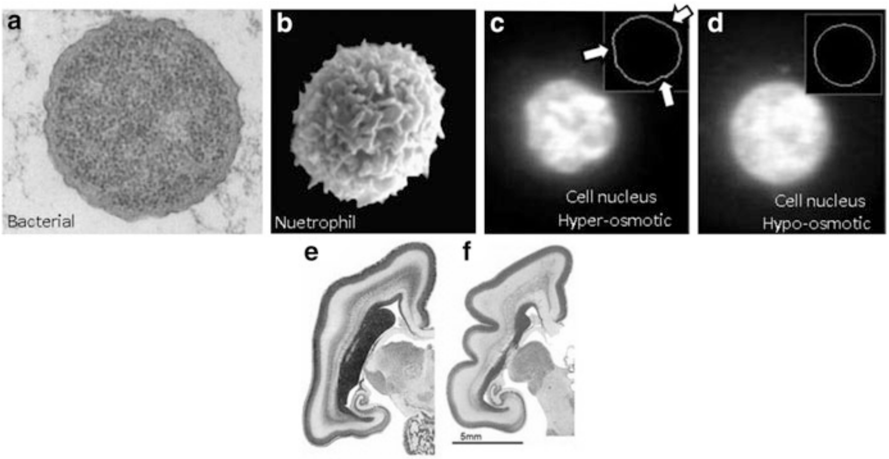

Fig. 2.7 The morphogenesis of some cells and tissues may be related to the wrinkling instability of nearly spherical shell/core systems. *For cells*: (**a**) Wrinkled bacterial cell owing to the relative shrinkage of the cytoplasm under hyper-osmotic pressure. (**b**) Wrinkled human neutrophil cell due to the relative expansion of the cell membrane surface area during cell growth or phagocytosis. (**c**) Wrinkled cell nucleus due to hyper-osmotic shrinkage, and (**d**) the wrinkles may disappear with the swelling of nucleoplasm under hypo-osmotic pressure. *The folding pattern of brain cortex*: the cross-sectional view of brain cortex shows that (**e**) during the early stage the surface is relatively smooth, and (**f**) during the later stage the wrinkled morphology is observed. Chen and Yin [76], reproduced with permission

by modifying the surfaces using focused ion beam [51]. External constraints may be applied, where a pre-patterned mold was held against the film as the buckles were formed, and the resulting pattern was quite stable after the removal of the mold [52]. The substrate may also be pre-strained [53], where silicon nano-ribbons bonded to a pre-stretched flat PDMS could generate wavy layouts upon releasing of the substrate strain (see Fig. 2.7g). Other than the discovered labyrinth, one-dimensional wavy and herringbone patterns in thin films, recently Chung et al. [32] demonstrated new types of wrinkles in a dendritic-like spoke pattern or in a target pattern consisting of concentric rings near defects in films (see Fig. 2.7h). Besides solid substrates, a recent work [54] reported a wrinkle-to-fold transition of buckled thin polyester film on water or soft gel substrates (see Fig. 2.7i).

In order to explain the formation mechanisms of various intriguing buckling patterns from theoretical aspects, Cerda and Mahadevan [55] proposed a generalized scaling law for the buckling wavelength and amplitude; Chen and Hutchinson [31, 56] showed that upon equi-biaxial compression of a film bonded to a planar semi-infinite compliant substrate, the herringbone pattern possesses less strain energy than its competitors and thus is more favorable. A more comprehensive discussion of the herringbone mode was recently given by Audoly and Boudaoud [57]. Huang et al. [58] analytically investigated the substrate thickness on the wrinkling wavelength and proved the feasibility of neglecting interface shear stress in theory.

By studying the kinetic buckling of elastic films on viscoelastic substrates, Huang [59] showed that both energetics and kinetics play important roles in determining the critical condition, the growth rate, and the wavelength. Through the control of anisotropic strains in films, the evolution and transition of stripes, herringbone, and labyrinth buckling patterns were simulated [60]. In a review, Genzer and Groenewold gave extensive examples of patterns achievable via film wrinkling and bridged that with skin wrinkles and possible ways of material characterization and fabrication.

2.2.3 Mechanical Self-Assembly (MSA) of Thin Film on Curved Substrate

The buckling characteristics of closed thin film (shell) on *curved* substrate (core) have important implications in the morphogenesis of quite a few fruits, vegetables, fingertips, animal skins, tissues, and cells [61–64] as discussed earlier. The intriguing wrinkling-like ordered patterns observed in these systems may be related to mechanics-driven buckling process owing to the mismatched deformation between the shell/film (e.g., skin of fruit or membrane of cell) and the underlying curved core/substrate (e.g., flesh of fruit or cytoplasm of cell), during which the curvature of the substrate may play a dominant role in shaping the distinctive overall appearance of quite a few natural and biological systems.

On morphogenesis, wrinkled cells are often observed in bacterial cells (Fig. 2.7a where the average wrinkle wavelength is about 100 nm) [65] and non-tissue cells such as human neutrophil cells (Fig. 2.7b) [66], macrophages, lymphocytes [67], and mast cells [68]. The wrinkled morphology may increase the surface area of the cell by more than 100 % [69], which may accommodate potential membrane expansion and spreading during extravasation and osmotic swelling. Other than the cell membrane surface wrinkles, recently similar wrinkled morphology was observed inside the cell, e.g., on the cell nucleus due to hyper-osmotic shrinkage, Fig. 2.7c [70]. In their work, Finan and Guilak [70] suggested that the nucleus wrinkles can be explained by the mechanical buckling of shell/core structure, where the contraction of the soft core (nucleoplasm) renders the stiffer shell (nuclear lamina) in compression to initiate the buckles. Under hypo-osmotic pressure, the swelling of nucleoplasm will make the lamina in tension and stretch the lamina into a smooth shape as shown in Fig. 2.7d [70].

Besides the cellular scale, the wrinkled morphology is also frequently observed at larger tissue or organ scales. The wrinkled brain cortex shown in Fig. 1.1c, d is a good example of wrinkling on curved substrates. The cross-section of the hemispherical cortex reveals the detailed information on the formation of the gyri (ridge) and sulci (groove) during development (see Fig. 2.7e, f) [71]. A number of hypotheses and models were proposed to explain how and why the cortex folds in a characteristic pattern from the biological, biochemical, and mechanical

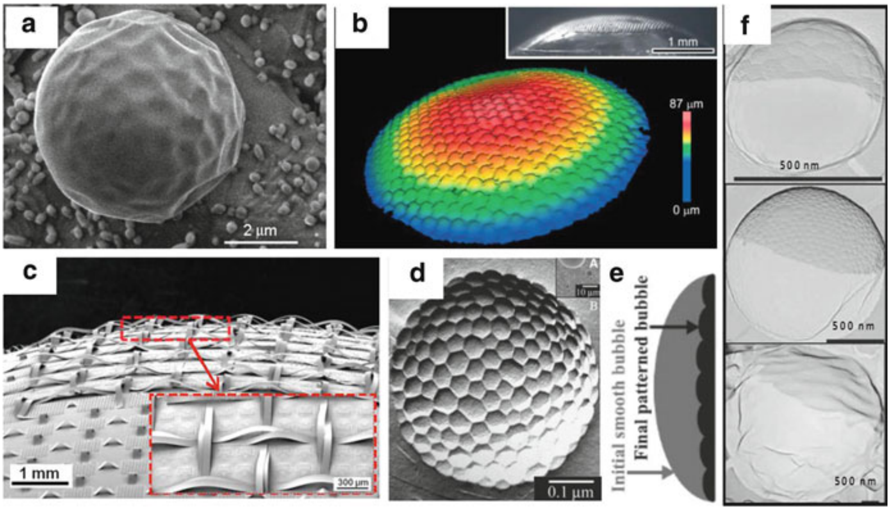

Fig. 2.8 Experiments of self-assembly on spherical shell/core systems. *Solid inorganic systems*: (**a**) Experiment of reticular pattern formed on a spherical system (SiO_2 film/Ag substrate). (**b**) Microlens arrays self-assembled on a hemispherical soft substrate using constrained local buckles. (**c**) Interconnected silicon ribbonlike photodetectors on a hemispherical elastomer substrate. *Instability patterns of microbubbles*: (**d**) Nanoscale hexagonal pattern self-assembled on a stable microbubble, which is in part due to (**e**) differential shrinkage-induced buckling of the bubble surface, and (**f**) the pattern can be strongly influenced by the bubble curvature. Chen and Yin [76] reproduced with permission

viewpoints [72–76]. Among them, Richman et al. [72] first proposed a mechanical buckling model where the cerebral cortex was modeled as a bilayer shell rested on a soft spherical core. The excessive growth of the shell relative to that of the core leads to the development of compressive stress in the shell, and the subsequent buckling may lead to the cortical folding.

Moreover, thanks to the rapid development of soft lithography which enables micro- and nanofabrication of multilevel thin-film devices with nonplanar geometries [77, 78], controlled 3D patterning on curved and/or closed substrates could significantly expand applications in biomedical engineering [79], optics [50], optoelectronics, and display technologies [80–82]. By investigating the undulation of a spherical Ag substrate/SiO_2 film system, Cao et al. [83] demonstrated an experiment of spontaneous buckling pattern formation on spherical substrates. Figure 2.8a shows an example with substrate radius $R = 3$ µm and at this relatively small R/t with $t = 150$ nm being the film thickness, reticular pattern was produced via spontaneous buckling. This serves an example of buckling self-assembly fabrication of true 3D structures at micro or submicron scales. To create 3D micro/nanopatterns and microstructures on curved substrates, Chan and Crosby [50] confined surface wrinkles in small local pre-patterned regions, and when such a technique was applied to a hemispherical surface, microlens arrays were self-assembled (Fig. 2.8b).

Recently, Shin et al. [84] assembled interconnected silicon ribbonlike photodetectors on a hemispherical elastomer substrate, and the ribbons were in buckled profiles owing to pre-stretch (Fig. 2.8c). It should be noted that in these experiments, the buckle features created were much smaller than the substrate radius of curvature; in other words, the versatile effect and potential of substrate curvature were not fully utilized to regulate the self-assembled buckles. Besides the aforementioned solid systems, buckling self-assembly was also demonstrated on spherical shell with hollow core microstructures (fluidic spherical shell/core microstructures), i.e., microbubbles. In a recent experiment on the nanopatterning of stable microbubbles, Dressaire et al. [85] created a nanoscale hexagonal interface pattern as shown in Fig. 2.8d through the shrinkage of the bubbles. By covering a surfactant layer on the surface of microbubbles, due to the differential shrinkage the initial smooth bubble buckled into a nano-hexagonal patterned one (Fig. 2.8e). Figure 2.8f further demonstrates the important effect of curvature on the surface wrinkling pattern, as the bubble radius was varied from 500 nm to 3 mm. Note that besides mechanical buckling, phase separation and other surface mechanisms may also underpin the pattern domains in microbubbles, and various pattern formations were reported including polygons, dendrites, beans, networks, etc. [86]. A recent review by Borden [86] has nicely summarized the nanopatterning on stable microbubbles, which have implications for biomedical applications.

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