Chapter 1
Mechanical Self-Assembly in Nature

Xi Chen and Jie Yin

Abstract Mother Nature provides unlimited inspirations of ordered patterns across vast scales: from the helical DNA and lipid bilayers at the submicron level, to the skin and tissue wrinkle at the millimeter level, to the ordered shapes in plants and animals at the meter level, and to the geological features at the mega scale. Many of these intriguing patterns are underpinned by mechanics-driven processes, including spontaneous buckling deformation.

1.1 Patterns and Morphologies in Nature

Over thousands of years of evolution, Mother Nature is able to self-assemble precise, differentiable, and intriguing morphologies on all scales from nano- to macro-size. For example, in cells, through bending and stretching deformation, the planar bilayer with phospholipids in solution can self-assemble into the liposome and micelles as shown in Fig. 1.1a. The self-folding of polypeptide chains into coiled proteins through bending and twisting is another famous molecular self-assembly example shown in Fig. 1.1b. At the tissue level, taking the development morphology of human's brain cortex for example, unlike a smooth kidney or

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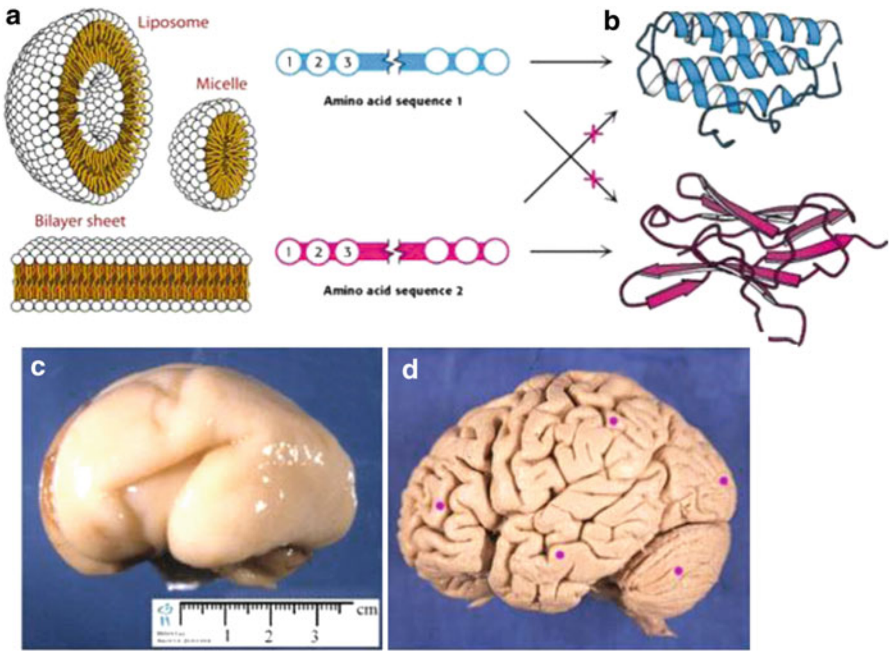

Fig. 1.1 Self-assembly in cells and tissues: (**a**) Three typical self-assembled structures of lipid bilayers in solution: liposome, micelle, and bilayer sheet. (**b**) Self-folding of polypeptide chains into proteins. (**c**) The smooth brain cortex of a fetus at 22 weeks. (**d**) The corrugated morphology of an adult human being's brain cortex. Yin [1], reprinted with permission

spleen, cerebral cortex is full of wrinkles and folds (see Fig. 1.1d). However, in the fetus period it is smooth (see Fig. 1.1c). At the early stage, as neurons continue to divide, grow, and migrate, the cortex folds and self-assembles a recognizable but unique pattern of bumps and grooves.

In the world of plants and fruits, Mother Nature creates more varieties of patterns and morphologies. Taking the different and intricate shapes found in leaves for example, leaves are seldom observed in perfect planar shapes; indeed, one of the most common morphologies is the saddle-like global appearance as shown in Fig. 1.2a (e.g., the rhododendron leaf). Moreover, a pear leaf often possesses a local wavy morphology along its edge (see Fig. 1.2b). Such a locally rippled edge is often found in the pond weed leaves; in addition, they often take the global curling and twisting topologies (see Fig. 1.2c).

A very special morphology is the tubular leaf shape found in rhododendron leaves in winter (see Fig. 1.2d), where the normally open and nearly planar leaf self-folds into a nearly closed tubular one as the temperature decreases. Other shapes including the doubly curled (see Fig. 1.2e) and spiral morphology (see Fig. 1.2f) are also found in leaves and tendrils. Figure 1.2g shows that most flowers have the similar wavy margins as leaves. A more complicated pattern is also observed in the sunflowers' heads and pine cones, where they have the beautiful and precise Fibonacci number patterns (see Fig. 1.2h, i).

Fig. 1.2 Different morphologies and patterns of leaves and flowers observed in nature. (**a**) Saddle-like rhododendron leaf. (**b**) A pear leaf with local wavy edge. (**c**) The global curled and twisted morphology of pond weed leaves. (**d**) The tubular morphology of rhododendron leaf (same as (**a**) except that the present one is in winter). (**e**) Doubly curled leaf. (**f**) Spiral morphology of a leaf. (**g**) Flowers with wavy margins in growth. Fibonacci number patterns in (**h**) sunflowers' head and (**i**) pine cones. Yin [1], reprinted with permission

Unlike balloons with a smooth surface, some fruits and vegetables distinguish themselves from others by possessing undulating surface morphologies as shown in Fig. 1.3.

For example, the small pumpkins such as acorn squashes show ten equidistant ridges running from the stem to the tip. Like the development of brain cortex, a pumpkin experiences a morphology transition from a smooth at newborn to the 10-ridged shape beyond a critical moment (and the overall ridged shape remains stable with continuous growth), as shown in Fig. 1.4. Similar 10-ridged morphology can also be found in the Korean melon (golden melon) and ridged gourd (or silk gourd, *Luffa acutanglula*). For another breed of big pumpkins, it often has about 20 ridges. Striped cavern tomatoes and bell peppers have 4–6 ribs to characterize their unique

Fig. 1.3 Examples of several natural fruits that exhibit distinctive buckle-like undulations as their global appearances. The number on the right corner shows the number of ridges. Chen and Yin [16], reproduced by permission of The Royal Society of Chemistry. The article in which this figure was originally published is located at the following link: http://pubs.rsc.org/en/content/articlelanding/2010/sm/c0sm00401d

Fig. 1.4 Morphology transition during the growth of a typical pumpkin from smooth surface to undulating morphologies. Yin [1], reprinted with permission

appearances. Different from the ribbed pattern, reticular cantaloupes demonstrate a netted pattern on their surface. In all these examples, the fruits/vegetables possess relatively stiff skin and relatively compliant flesh, and the overall shape is approximately spheroidal. Another special case is the wax apples, whose overall shape is conical and they exhibit a beautiful skin wrinkle-like ridged morphology on their surfaces. Note that for the same breed of fruits, the ridge number remains essentially unchanged despite their different sizes. Interestingly, similar ridged patterns can be also found in butterfly, bollworm and tobacco budworm eggs, dehydrated fruits and nuts (e.g., almond and prune), animal skin, tissues, etc.

Other than the fascinating self-assembled patterns and morphologies in biological systems, Mother Nature's strong power is witnessed even at the macroscopic geological systems. In many polar and high alpine environments, striking

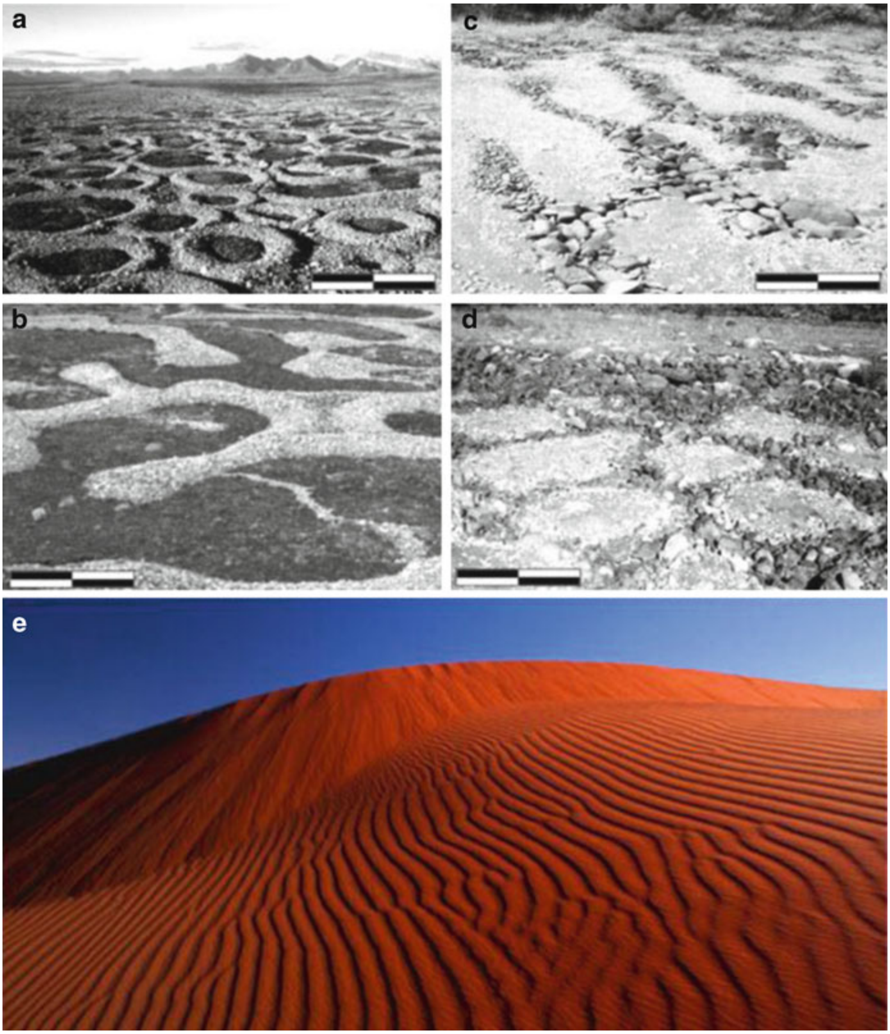

Fig. 1.5 Forms of sorted patterned ground: (**a**) sorted circles and (**b**) sorted labyrinths, (**c**) sorted stripes, (**d**) sorted polygons (full scale bar: 1.0 m). (**e**) Rippling sand dunes under wind. Yin [1], reprinted with permission

circular, labyrinthine, polygonal, and striped patterns of stones and soil are spontaneously formed through self-organized processes (see Fig. 1.5a–d) [1], which constitute one of the most striking suites of geomorphic patterns. In sand dunes, the self-assembled meandering rippled pattern often observed in deserts is another beautiful scene in deserts [2], where the shape of ripples keeps across several thousand meters (see Fig. 1.5e).

1.2 Underlying Mechanical Mechanisms for Self-Assembled Pattern Formation

The self-assembled patterns and morphologies in Nature have fascinated and challenged both biologists and geomorphologists for decades [1, 3, 4]. For instance, the mechanisms underpinning the formation of fascinating shapes and patterns in plants have evoked scientists' interests for centuries [4]. While there is no doubt that biological and genetic factors significantly influence the morphogenesis, the active role of physics and mechanics should not be underemphasized, as stated by D'Arcy Thompson in his classical book on growth and forms [4],

> Cell and tissue, shell and bone, leaf and flower, are so many portions of matter, and it is in obedience to the laws of physics that their particles have been moved, moulded and conformed.

Thompson's argument on the inevitable interactions between physics and biology has been evidenced by recently increasing works, where the important role of mechanical force is found in the regulation of plant morphology [5, 6], cell growth and cell differentiation [7], and tissue morphogenesis [8], among others. Recently Hamant et al. [6] showed that mechanics plays a key active role in the development of plant organs, where the mechanical stress on plant tissue during growth controls the precise organization of a major structural element in the plant cell. By applying the growth hormone auxin to the edge of an eggplant leaf to cause a local expansion, Sharon et al. [9] showed that the growth stress makes the normally flat leaf buckle into a wavy one. Similar studies on the control of crinkly leaves were conducted by Nath et al. [10] even at the genetic level and they showed that flat leaves become crinkly in *Antirrhinum* after the *CIN* gene mutant, where leaves of *cin* cause excess growth in marginal regions and the resulting growth stress may lead to a curved surface. By investigating the undulation of a spherical Ag substrate/ SiO_2 film system, Li et al. [11] successfully reproduced the Fibonacci number patterns in experiment that are similar to those observed in sunflowers' head, pine cones, and other plants, which suggested that mechanical force may be a driving force for some plant pattern formation.

For fruit morphology, from the mechanics point of view, the transition process from a normally smooth topology to a global undulating one (e.g., Fig. 1.3) may correspond to the occurrence of mechanical instability. Upon instability, the system will spontaneously choose the buckling mode with the lowest energy, often with an ordered self-assembled undulating pattern. Likewise, one may suspect that when the accumulated growth stress in the fruit arrives at the threshold, instability may occur and the spontaneous buckling pattern may underpin the distinctive morphologies observed in nature. This mechanical process may set a template for the subsequent and complex biological and biochemical processes (e.g., cell growth and differentiation) which may help to stabilize the distinctive patterns [12].

1.3 Bio-Inspired Self-Assembled Micro/Nanofabrication

Other than the spontaneously formed patterns in nature, in engineering, patterning has been developed as a technique for several decades to create functional and desired patterns and structures at the micro- and nanoscales. For example, in the field of microelectromechanical systems (MEMS), the traditional micro/nanofabrication techniques based on photolithography [13, 14] have been highly developed to fabricate micro- and nanopatterns and structures in the past 30 years [15]. The conventional photolithography approach uses UV light to transfer mode patterns to light-sensitive chemical photoresists on the underlying films or substrates, and after chemical etching patterned structures with high accuracy are produced. However, these methods are intrinsically expensive, time-consuming, effective to only a set of photoresists, and not directly applicable to nonplanar surfaces [16]. One of the central drawbacks is its limitation of fabricating small-aspect-ratio microstructures owing to the difficulty in deep etching methods [17]. In recent years, the *Lithographie*, *Galvanoformung*, *Abformung* (LIGA) technique has been developed to fabricate high-aspect-ratio microstructures to overcome the limitation [18]; however, it is low-efficient and highly expensive. The other main limitation of these traditional techniques is that the produced patterns and structures are inherently two dimensional (2D) as a result of the wafer-based fabrication methods. It is extremely challenging to create truly three-dimensional (3D) microstructures (e.g., on curved substrates) using lithography-based techniques. Thus the fabrication of 3D micro/nanopatterns and structures with high efficiency and low cost calls the need of revolutionary alternative approaches.

Among the conceptually new strategies offering possible routes to both small features and low costs, self-assembly involving thin films and multilayers has become the new cornerstone [16, 19]. The driving force for the thin film self-assembly can be the chemical interaction between film and substrate or physical forces such as magnetic, capillary, dispersion, and entropic interactions [16, 19]. Recently mechanical self-assembly of stiff film on planar substrate system driven by mismatched deformation has received wide applications in engineering [20], which offers a cost-effective and complementary solution to overcome the aforementioned challenges. The rich parameters that can be manipulated during the mechanical self-assembly process, including the material properties of film and substrate, substrate curvature, material and geometry gradient, mismatched stress, and time-dependent properties, etc., provide unlimited possibilities for the creation of varieties of functional 2D and 3D patterns and structures, with wide potential applications in biomedical engineering [21], optics [22], optoelectronics and display technologies [23, 24], among many others.

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