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Review

Versatile Rolling Origami to Fabricate Functional and Smart Materials

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SUMMARY

Fabrication methods inspired by origami are of great potential in 3D micro-/nanostructure construction. Typically, it exploits the bending of planar nanomembranes, including both partial deformation for folding and global deformation for rolling. Here, we focus on rolling origami technology, which is a crucial route to fabricate cylindrical micro-/nanostructures. The versatility of rolling origami is discussed, with a focus on material choice, flexible designability of rolled-up structures, and a wide range of demonstrated applications. Reconfigurable properties of rolled-up micro-/nanostructures are highlighted when rolling origami is combined with smart materials. Finally, current limitations and the next steps toward idealized rolling origami fabrication are outlined.

INTRODUCTION

Origami, as a kind of talented paper art, generally refers to folding planar paper into various stereo artifacts. With increasing demands for three-dimensional (3D) microdevices, similar concepts have been applied to micro- and nanoworlds to transform patterned planar nanomembranes into 3D shapes and have become an important part of 3D micro-/nanostructure construction.[1,2] In a broad sense, origami is based on the bending of materials, which can be divided into two phenomena.[3,4] One is localized bending designed with rigid and soft parts, i.e., folding, which is regarded as traditional origami.[5] The other is bending in the whole body, usually resulting in the rolling of planar material.[6] To avoid ambiguity, we use the term "rolling origami" to clarify the latter transformation process through which planar materials are rolled into 3D structures.

Rolling origami was established from 2000 onward.[7,8] Introducing vertical strain gradient into nanomembrane system opens up a new era of 3D microstructure design. Upon release from substrate by etching the sacrificial underlayer, these strained nanomembranes tend to force themselves into cylinder structures to stabilize their elastic energy,[9] leading to free-standing 3D forms. Since then, the development in rolling origami has brought about a large variety of 3D microdevices spanning optical and electrical areas to biological applications. In this review, we focus on this strain-induced origami of nanomembranes to introduce this powerful fabrication method with exceptional versatility in materials and structures, as well as novel properties and applications of 3D cylinder microstructures fabricated from rolling origami (Figure 1). We summarize several systems of rolling origami divided by sacrificial layers (a layer that assists rolling), especially focusing on their compatibility with materials. Then, we discuss rolling strategies for structure design with desired 3D geometries, followed by typical functions of different kinds of functional nanomembranes. In the end, a discussion on the concept and applications of rolling origami with smart materials is given, in which rolled-up microstructures change their shapes

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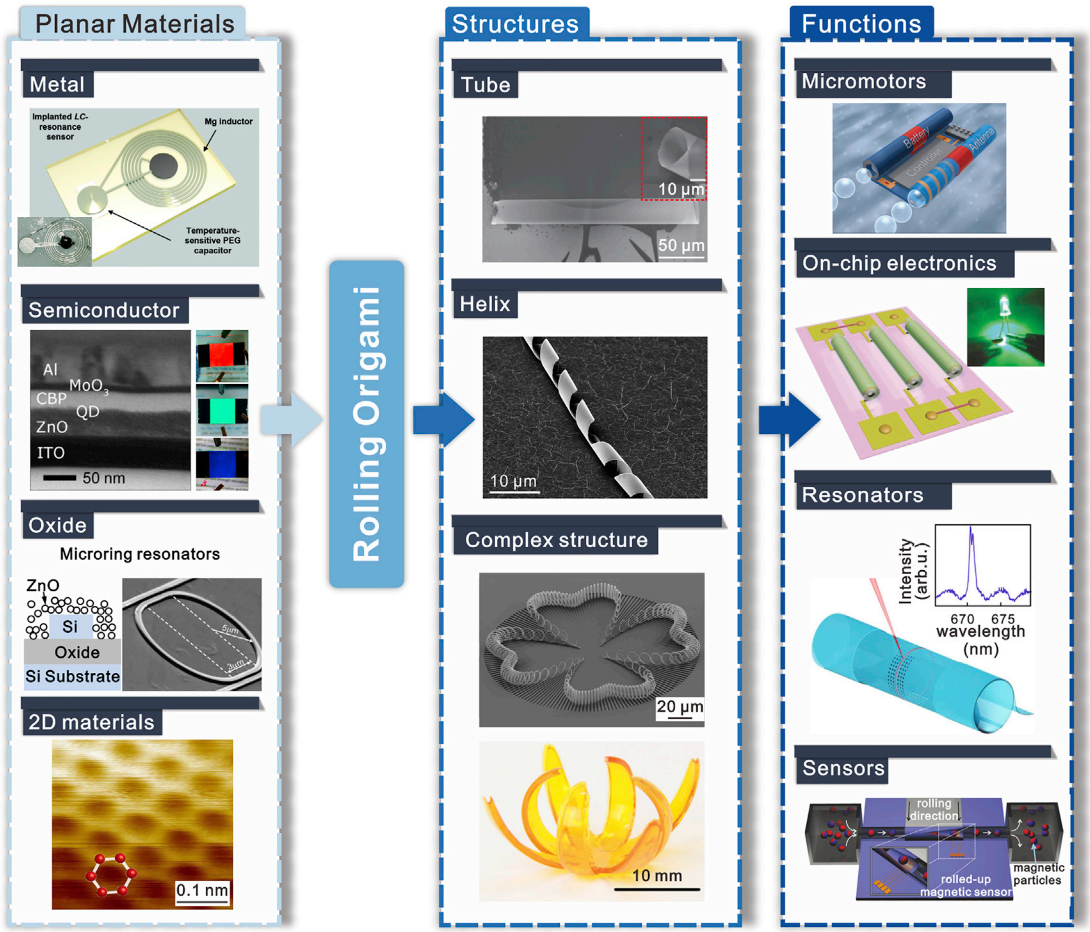

Figure 1. Rolling Origami Transforming Planar Materials into 3D Microstructures for Realization of Different Functions

Through rolling origami, films or nanomembranes made of different materials (metals, semiconductors, oxides, and 2D materials)[10–13] are able to be transformed into 3D microstructures as tubes, helices, and others.[14–17] Various applications, including micromotors, on-chip electronics, resonators, and sensor, are achieved on rolled-up microstructures.[18–21] Copyright, 2020, Wiley-VCH. 2012 American Chemical Society. 2010, OSA. 2007, The National Academy of Sciences of the USA. 2020, Chines Institute of Electronics. 2019, American Chemical Society. 2006, American Chemical Society. 2018, American Chemical Society. 2011, The Royal Society of Chemistry. 2019, The Author(s), CC BY 4.0. 2018, American Chemical Society. 2011, American Chemical Society.

under external stimuli. And we put forward the next step to develop "real" rolling origami, through which all kinds of rolled-up microstructures can be formed by advanced pattern design and precise rolling control, accompanying with low costs and extended functions.

Theory of Rolling Origami

Before reviewing the realization of rolling origami through nanotechnology, we would like to give a brief introduction about the mechanism and theory related to rolling origami. Another review gives a comprehensive summary of related theory in the formation of rolled-up tubes and helices.[9] Considering the thickness of nanomembrane usually ranging from several to tens of nanometers, it is difficult to make a soft nanomembrane roll up and maintain its shape directly by an external force. Instead, the force to roll up a free nanomembrane generally comes from the nanomembrane itself, that a strain parallel to the surface is established. Additionally, strain inside nanomembrane requires a difference in the normal direction, and this strain gradient can force released nanomembrane to bend and roll.

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Unfortunately, rolling is not the only result for a released nanomembrane with an established internal strain gradient. A failure is found in experiments that sometimes released nanomembrane forms wrinkled shapes rather than rolled-up structures. This phenomenon was theoretically discussed by Cendula et al.[22] They calculated the bending and wrinkling energy in a bilayer system with determined strain and applied energy minimization principle to determine the outcome shape of released nanomembranes. They found that wrinkles prefer to appear in the system with small strain gradient. Thus, creating sufficiently large strain gradient in the nanomembrane system is an important aspect to obtain successful rolling origami.

Furthermore, elastic mechanics provides a reliable theory and model to guide the formation of rolled-up microstructures. A general and useful formula to calculate the curvature of rolled-up nanomembrane is given by Nikishkov.[23]

$$K = \frac{3\sum_{i=1}^{n} E_i' t_i (z_i + z_{i-1} - 2z_b)\left(c - v_i' \varepsilon_i^0\right)}{2\sum_{i=1}^{n} E_i' t_i \left[z_i^2 + z_i z_{i-1} + z_{i-1}^2 - 3z_b (z_i + z_{i-1} - z_b)\right]} \quad \text{(Equation 1)}$$

This formula is applied to estimate the curvature in the nanomembrane system with n layers counting from bottom to top. Properties of each layer include equivalent Young's modulus $E_i^{'}$ and Poisson ratio $v_i^{'}$ under the plane strain assumption. Geometric parameters include the thickness of each layer t_i and the location of upper surface z_i ($z_0 = 0$, $z_1 = t_1$, $z_2 = t_1 + t_2$, …). z_b and c represent the location of neutral plane and uniform strain after relax, respectively. Also, simplified formula can be used in some specific situations like bilayer system and layers with relaxed part.[24] More than analytical calculation, modeling method is developed to explain or predict rolling behavior. One of the most representative works is reported in 2014.[25] A quasi-static modeling was invented to simulate the rolling process during releasing step by step. This method gives a good prediction in the resultant microstructure from rolling origami, especially for those with complex pattern designs that cannot be regarded as a 2D question.

Material Compatibility

The compatibility with different materials is one of the most important issues for a 3D micro-fabrication technology to be widely applied, especially when the technology is highly integrated with various functional blocks. Over the past few decades, materials permitted in rolling origami have been developed from a limited range of materials to extraordinarily diversified material systems. A standard material system of rolling origami consists of a bottom substrate, a sacrificial layer in the middle, and functional layers (including strained layers) on the top. The sacrificial layer, deserving the name, is doomed to "sacrifice" in order to release strained layers for accessible rolling.

In most cases, when a sacrificial layer is settled, the range of functional layers is determined due to the requirement of different etching properties in the etchant of sacrificial layer. To trace the evolution of material systems in rolling origami, fabrication processes are introduced in this chapter according to sacrificial layers for their great relevance to functional layers. We introduce material systems from inorganic to organic sacrificial layer, followed by the material layers used in newly developed rolling origami without sacrificial layers. A brief summary of materials and techniques applied in rolling origami is given in Table 1.

Inorganic Sacrificial Layer

Inorganic materials are used as sacrificial layer since the earliest realization of rolling origami back in 2000.[7] Until now, inorganic sacrificial layers, including III-V

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Table 1. Summary of Materials and Techniques Applied to Rolling Origami

Rolling Origami with Sacrificial Layers

Sacrificial Layer Material	Release Method	Layer System	Typical Reference
AlAs	HF-based solution	GaAs/InAs	7,26
		InGaAs/GaAs/Cr, InGaAs/GaAs/Thiol	
		$InGaAs/Fe_3Si$	
AlGaAs	HF-based solution	GaAs/AlGaAs/GaAs/AlGaAs/InGaAs	27
Undoped-Si	NH_4OH solution	Si/SiGe	28
Si	HNO_3 and HF solution	AlN/GaN	29
SiO_2	HF solution	Si/DLC	30
Ge	water	$Au/Cr/TiO_2$	31–33
	H_2O_2 solution	SiN_x/SiN_y	
	XeF_2 vapor (dry)	$Al_2O_3/Ni/Cu/SiN_x/SiN_y/Al_2O_3$	
GeO_x	H_2O_2 solution	Pt/RuO_2	34,35
		$TiO_2/Pt/RuO_2$	
		$Pt/Py/Pt/Al_2O_3/SiO/SiO_2$	
Al	KOH	Au/Ti	36
Cu	$HClO_4$ and $Ce(NH_4)_2(NO_3)_6$ solution	SiO/SiO_2, $SiO/SiO_2/Fe$, $SiO/SiO_2/Pt$	37
		TiO_2,Ti	
		Si/TI	
Photoresist	acetone solution	Pt, Pd/Fe/Pd	38
		TiO_2, ZnO, Al_2O_3	
		Si_xN_y, Si_xN_y/Ag, DLC	
PMMA	heating, 180°C	$SiO/TiO_2/Au$, $SiO/TiO_2/Ag$	39
		$SiO/TiO_2/Pt$	
PAMS	heating, 300°C	Ge	40
		Y_2O_3/ZrO_2	
		Ti/Cr/Pt, Ti/Fe	
LA-PAA metal organic layer	sodium DETPA solution	NiFe/Cu/NiFe	41,42
		$Al_2O_3/IGZO/Ti/Au/Al_2O_3$	
Methyl cellulose	water	$Al_2O_3/(Ni/Cr)/Al_2O_3/(Ni/Cr)$	43
PS/P4VP	heating, 500°C	Au, Ti, Au/Ti	44

Rolling Origami without Sacrificial Layers

Mechanism	Layer System	Typical Reference
Increasing strain gradient by swelling	PSI/PCL	50
Increasing strain gradient by thermal expansion of FC	Ti/Cr, $Ti/Cr/Al_2O_3/Cr/Al_2O_3$	51
	$Ti/Au/TiO_2/Cr$	
Decreasing adhesion by ethanol solution	2D TMD	52
Decreasing adhesion by droplet	Ti/Cr	53
	SiO/SiO_2	
	Au/SiO/Fe, Au/SiO/Fe/Ag	

semiconductor, silicon (Si), silicon oxide (SiO_x), germanium (Ge), and its oxide (GeO_x), are still widely applied to suit the needs of fabrication and working circumstances with some functional materials. These sacrificial layers are etched away in corresponding etching solutions. Generally speaking, the etching rate of the sacrificial layer with a chosen etching solution should be much faster than that of upper layers, which protects strained nanomembranes to a maximum extent. Therefore, material restriction exists in the nanomembrane system because not all materials are tolerant to the etching solution. Other than sacrificial layers like III-V semiconductors and Si-related materials, research of inorganic sacrificial layers is also focused on reducing the selectivity of upper layers and developing mild/environmental-friendly etching solution.

In the early development of rolling origami, III-V semiconductor sacrificial layers, together with upper layers, are deposited by molecular beam epitaxy (MBE). This fabrication process allows the formation of monocrystalline nanomembranes in high quality, leading to novel and outstanding physical properties. As shown in Figure 2A, Prinz and his team devoted themselves to rolling 3D III-V semiconductor structures using AlAs sacrificial layers.[7] InAs/GaAs bilayer is deposited on the AlAs sacrificial layer, where the strain gradient of the bilayer is determined by the lattice mismatch as $\Delta a/a$ = 7.2%. The interatomic force of F_1 in the InAs layer tends to increase the interatomic distance although F_2 in the GaAs layer acts oppositely. So, the forces F_1 and F_2 produce a moment of force M to roll the bilayer when the AlAs layer is etched away by etchant based on hydrogen fluoride (HF). Similar to AlAs, other semiconductors, including AlGaAs,[45] AlSb,[46] AsSb,[47] and CdTe[48] are utilized as sacrificial layers. Figure 2B presents an instance in which AlGaAs serves as a sacrificial layer with upper layers consisting of strained bilayer and high-quality semiconductor quantum wells.[27] It is concluded that the utilization of MBE highly improves the quality of deposited nanomembranes, paving the way to study advancing properties and potential applications of rolled-up microstructures. But also, its high cost hampers it to be applied in a general situation. Besides, benefiting from the strain of lattice mismatch, the strain gradient in layers can be precisely defined and altered according to the component of each layer. We may find that the materials of the strained layers are limited in III-V semiconductors, and most of the time, strained layers work as functional layers. But when we need other additional functional materials on top of the strained layers, the choice is beyond the boundary of semiconductors for the additional layers being no more single crystalline, such as metals, Al_2O_3, and organic 1-hexadecanethiol.[26]

Moreover, silicon-based sacrificial layers are developed, as selectively etching of such materials has been widely used in the semiconductor industry. Prinz et al.[28] take undoped Si as a sacrificial layer and NH_4OH solution as corresponding etching solution, utilizing MBE to grow Si/SiGe lattice-mismatched bilayers. Fortunately, MBE is no longer a necessity in Si-based fabrication procedure; thus, the range of permitted materials is extended other than semiconductors. And also, the strain can be introduced by the disparity of the thermal expansion coefficient. Figure 2C exemplarily shows an ultrathin, porous AlN/GaN bilayer deposited by metalorganic vapor phase epitaxy on Si substrate.[29] Here, when the layer thickness is in the range of 20–35 nm, the rolling strain is caused by zipping effect, which is induced from the AlN island formation during growth. Si sacrificial layer under AlN is removed by a mixture etchant consisting of HNO_3 (69%) and HF (50%) with a volume ratio of 10:1. Figure 2D shows another example, in which SiO_2 is used as a sacrificial layer, with the inherent strain in the diamondlike carbon (DLC) layer to assist the rolling of DLC/Si bilayer.[30] Here, SiO_2 sacrificial layer is a built-in nanomembrane with

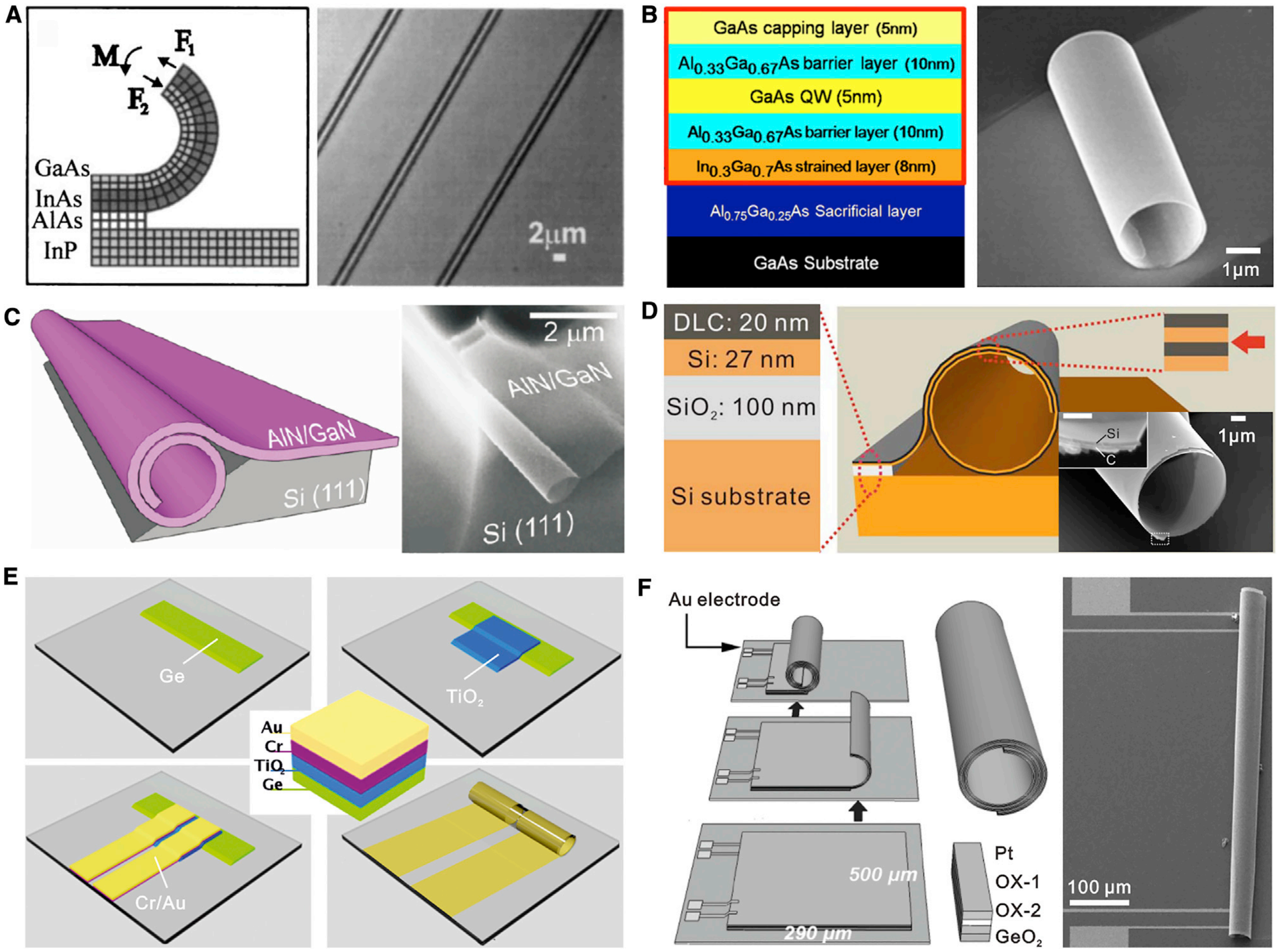

Figure 2. Material Compatibility of Rolling Origami with Inorganic Sacrificial Layers
(A) GaAs/InAs bilayer microtube constructed from etching AlAs layer in HF solution.[7] Copyright, 2000, Elsevier.
(B) Self-rolled-up microtube released from $Al_xGa_{1-x}As$ layer.[27] Copyright 2010, American Institute of Physics.
(C) AlN/GaN nanomembrane released by etching Si in HNO_3/HF mixture.[29] Copyright, 2009, American Chemical Society.
(D) Fabrication of C/Si microtube on SiO_2 layer with HF wet etching.[30] Copyright, 2009, American Institute of Physics.
(E) Self-rolling with patterned Ge sacrificial layer, dissolved in water.[31] Copyright, 2014, American Chemical Society.
(F) Nanomembrane released from GeO_2 in H_2O_2 solution.[34] Copyright, 2017, Wiley-VCH.

thickness around 100 nm in Si-on-insulator (SOI) wafer, which is eliminated by HF solution (49%). It is obvious that, with the employment of Si and its oxide as sacrificial layers, more explorations are done to increase the diversity of strained layers as well as functional layers. Taking advantage of this silicon-based fabrication process, the complexity of rolled-up microstructures highly increases due to its compatibility with conventional planar technology. And rolling origami is able to meet the demands of on-chip microdevices, which is considered as one of the most desired issues in 3D microstructures.

In above rolling origami systems, the etching of sacrificial layers commonly occurs in highly toxic and dangerous solutions as HF. This requirement is obviously not acceptable and may bring harmful contaminants in biological situations and investigations. Instead, some inorganics diluted in a mild solution can serve as sacrificial layers with little effect on upper nanomembranes. Oliver and his group members selectively etch Ge sacrificial layer in water to release strained multilayer nanomembranes, as shown in Figure 2E.[31] They also successfully roll up nanomembranes by removing GeO_2 in 1.0% H_2O_2 (Figure 2F).[34] The concentration of H_2O_2 determines the time of releasing. For example, GeO_x layer is immersed in 0.03% H_2O_2 at 50°C for 4 h and deionized water for 2 days to complete the rolling origami process.[35] To shorten the time of etching process of GeO_x and Ge layer, one simple method is to increase reaction temperature and the concentration of H_2O_2. It is reported that, in 2% H_2O_2 at room temperature, the etching speed of Ge sacrificial layer is ~9.72 nm/s, much slower than 480 nm/s in 30% H_2O_2 at 70°C.[32] And dry etching technique with XeF_2 vapor is reported to etch Ge sacrificial layer, producing a rolling speed of ~750 μm/min. This speed is 500 times faster than wet-etching method and thus enables large-scale production.[33] Different from Si-based ones, Ge-based sacrificial layers are environmentally friendly and make the device compatible for end-used bioapplications, like cytometry, cell sorting, and biosensing.

Organic Sacrificial Layer

Complex functional material consideration is usually required when taking inorganic material as a sacrificial layer in the fabrication process, concerning the different etching rates of etching solution to the sacrificial layer and deposited nanomembranes. The emergence of organic sacrificial layers simplifies the selection of inorganic functional materials, realizing a versatile approach of rolling origami. Organic sacrificial layers become a vital part in rolling origami as their ability in increasing the feasibility and applicability of rolling origami in real application scenarios. Representative organic sacrificial layers are shown here to reveal their recent developments for processing improvements and more compact and dedicated structures.

In 2008, Mei et al.[38] proposed a grand rolling origami method by applying photoresist as sacrificial layer, as shown in Figure 3A. With photoresist sacrificial layer, rolled-up microstructures in a series of material combinations are obtained, including Pt, Pd/Fe/Pd, Al_2O_3, and Si_xN_y. Notably, the patterning process is directly completed by photolithography of the sacrificial layer. After deposition of nanomembranes, materials deposited on the patterned photoresist are released by immersion in acetone. The etching solution of photoresist has scarce effect on upper inorganic layers. Thus, a broad range of materials and their combinations is available in rolling origami fabrication. In addition, compared with conventional steps of rolling origami consisting of depositing, shaping, and releasing nanomembranes, the procedure of rolling origami is simplified by the combination of depositing and shaping steps using photoresist. Since then, this simplified method has demonstrated a series of microstructures functionalized in various applications ranging from photonics to biophysics.[49]

What if the material system including polymer functional layers or complex 3D microstructures needs multi times of photopatterning? In these cases, chemical incompatibility would occur when using an organic sacrificial layer relying on single-step lithography. Besides, various polar and nonpolar solvents, especially those with strong acidic and alkaline solutions, could devastate the organic sacrificial layer during repeating lithographic steps. In turn, the etchant solution could do harm to the polymer upper layers when removing organic sacrificial layer. To solve these problems, Karnaushenko et al. developed a new polymer platform, as shown in Figure 3B.[41] The sacrificial layer applied here is imide- and acrylic-based polymer (metal-organic sacrificial layer, La-PAA), which is not only photo-patternable but also chemically and thermally stable. A strained bilayer is separately designed on the La-PAA sacrificial layer, which is made of a hydrogel layer with swelling property

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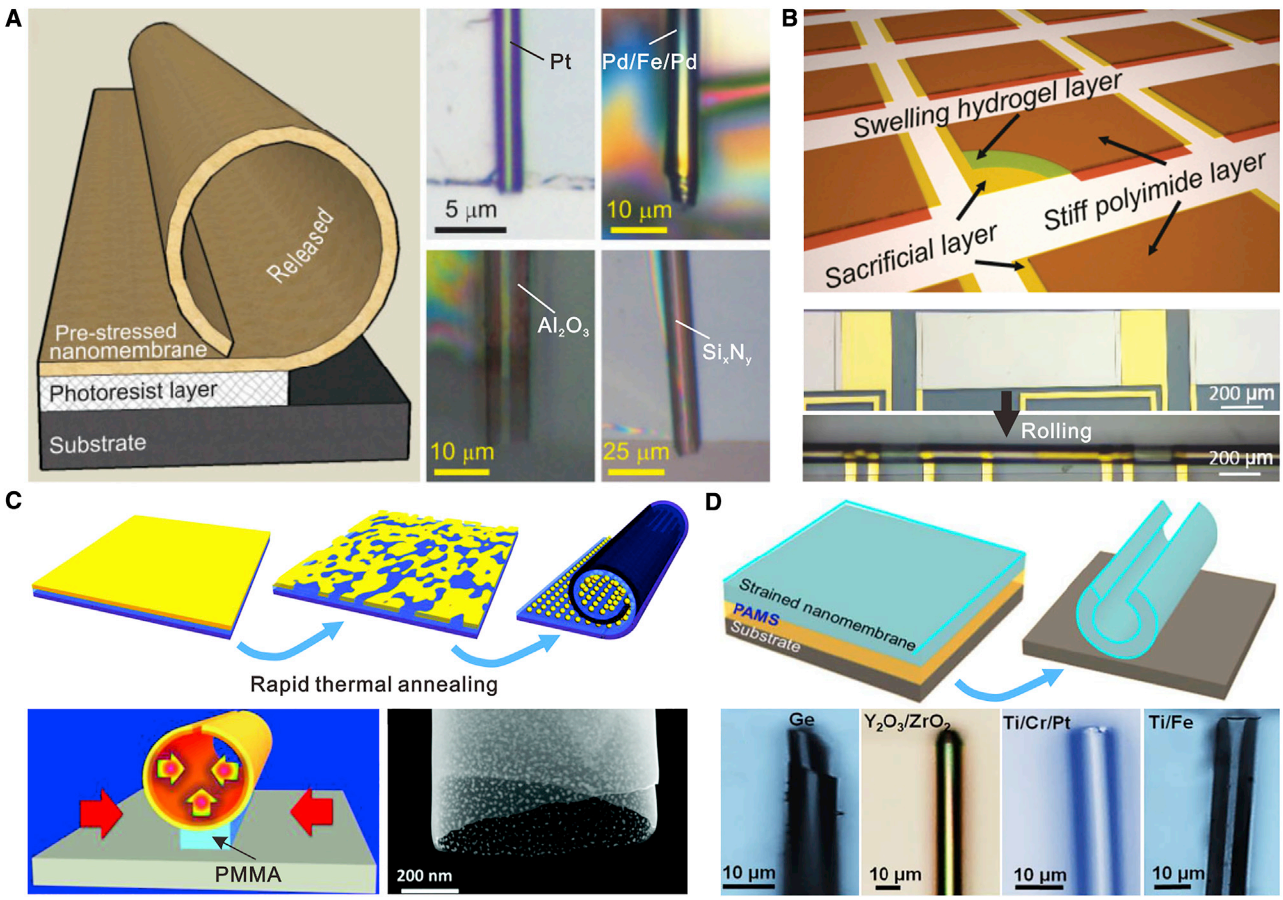

Figure 3. Material Compatibility of Rolling Origami with Organic Sacrificial Layers

(A) Various inorganic microtubes fabricated on photoresist etched by acetone.[38] Copyright, 2008, Wiley-VCH.

(B) Complex multilayer nanomembrane fabricated on metal-organic sacrificial layer with mild etching requirement.[41] Copyright, 2015, Wiley-VCH.

(C) Dry etching of PMMA layer with rapid thermal annealing. Internal metal layer transforms into nanoparticles to provide external driven force.[39] Copyright, 2013, Wiley-VCH.

(D) Assembling of different rolled-up nanomembranes through polymer pyrolysis.[40] Copyright, 2019, IOP Publishing.

and a stiff polyimides layer. The etchant solution here is 0.5 M sodium diethylenetriaminepentaacetic acid (DETPA) water solution. During the releasing process in DETPA, the hydrogel layer absorbs water and generates mechanical stress paralleled to the layer, leading to upward force to achieve rolling behavior. The key advantage of La-PAA sacrificial layer is that multiple functional layers are shaped successively into different functional elements and integrated into a single fabrication run, enabling rolling origami with complex microelectronics fabrication technology (e.g., CMOS process). Similar to photoresist sacrificial layer, this platform based on La-PAA sacrificial layer uses standard equipment in Si integrated technologies as lithography and deposition methods. And the biocompatibility of the La-PAA-based rolling origami method has also been demonstrated.[42]

For the most part, etching process of sacrificial layers is based on wet-etching methods that are compatible with Si-integrated technologies. After the wet-etching process, additional liquid-removing treatments are almost inevitable to get independent rolled-up microstructures. This may cause collapse of these hollow 3D structures due to the surface tension during the evaporation of liquid, reducing the yield of microdevice production. Therefore, critical point drying (CPD) is always applied to dry samples for preventing this phenomenon. On the other hand, this issue is circumvented by dry-releasing methods, through which no liquid takes part in the etching process. A few papers adopt dry-releasing methods for a potential solution to future industrial production. One example is shown in Figure 3C. Li et al.[39] applied polymethyl methacrylate (PMMA) as sacrificial layer and removed it with heating treatment at 180°C in a nitrogen atmosphere. With rapid thermal annealing, the PMMA sacrificial layer is burnt away. At the same time, metal layers with several nanometer thicknesses are reorganized into nanoparticles, generating additional strain due to surface tension, resulting in the formation of rolled-up nanostructures with a diameter in hundreds of nanometers. In Figure 3D, pyrolysis of polymer is taken into consideration for rolling origami, waiving wet etching and accompanied CPD step.[40] The rolling process is triggered at 300°C, at which temperature the poly-α-methylstyrene (PAMS) sacrificial layer is degraded into gaseous monomer molecules. Through this method, a set of inorganic rolled-up microstructures are successfully fabricated. The dry-etching releasing methods create possibilities for rolling origami toward future mass production in a simple procedure, as well as increasing the stability of structures by avoiding additional drying treatment. Unfortunately, by now, dry releasing methods are not as mature as wet etching in structure and function design. We look forward to an optimal polymer sacrificial layer with proper pyrolysis temperature to permit the use of various materials, as well as precise control with localized heating methods.

No Sacrificial Layer

Working as a bridge to attach functional layers onto a supportive substrate, the sacrificial layer "dies" in the releasing process, resulting in a full strain release of upper layers. Considering that the sacrificial layer is used to peel off nanomembranes from the substrate, there is no need to insert it if deposited nanomembranes are spontaneously delaminated from substrate. When the strain gradient is larger than the adhesive force, upper layers can delaminate and roll into pre-designed 3D microstructures. In turn, the layers stay on the substrate if adhesion is greater than the strain. Unfortunately, these two forces are usually fixed as the substrate and nanomembrane are determined; thus, it is hard to control delamination behavior after deposition. But there are several reports with clever design to tune the force relationship for the realization of rolling origami, which follows the strategy as increasing the strain inside or reducing the adhesion. Therefore, no sacrificial layer and additional etching process are required.

In this part, we show several examples in which the delamination occurs via increasing the strain inside nanomembranes. Figure 4A exhibits a method using two polymer layers with distinct swelling ability in certain solution.[50] When cross-linked polysuccinimide (PSI)/polycaprolactone (PCL) bilayers are immersed in a physiological buffer (pH 7.4), the bottom PSI layer starts to hydrolyze and swell slowly, tending to stretch its body. On the contrary, the top PCL layer remains water insoluble, resulting in an additional strain gradient perpendicular to the nanomembrane. Then, this strain gradient makes nanomembrane peel off from the substrate and roll into microtubes. Additionally, Figure 4B presents a delamination process using an amorphous fluorocarbon (FC) layer upon thermal treatment.[51] FC is an anti-adhesive polymeric layer between nanomembranes and the substrate, which has gained thermal expansion differences compared with inorganic layers. With the rising temperature, expansion in the FC layer leads to two delamination circumstances as mechanism I and II. The mechanism I refers to the detachment of nanomembrane from FC layer although mechanism II refers to the delamination of the FC layer from substrate. Whether the FC layer is rolled up or not depends on the adhesion between the substrate and the FC layer compared to that between the FC layer and

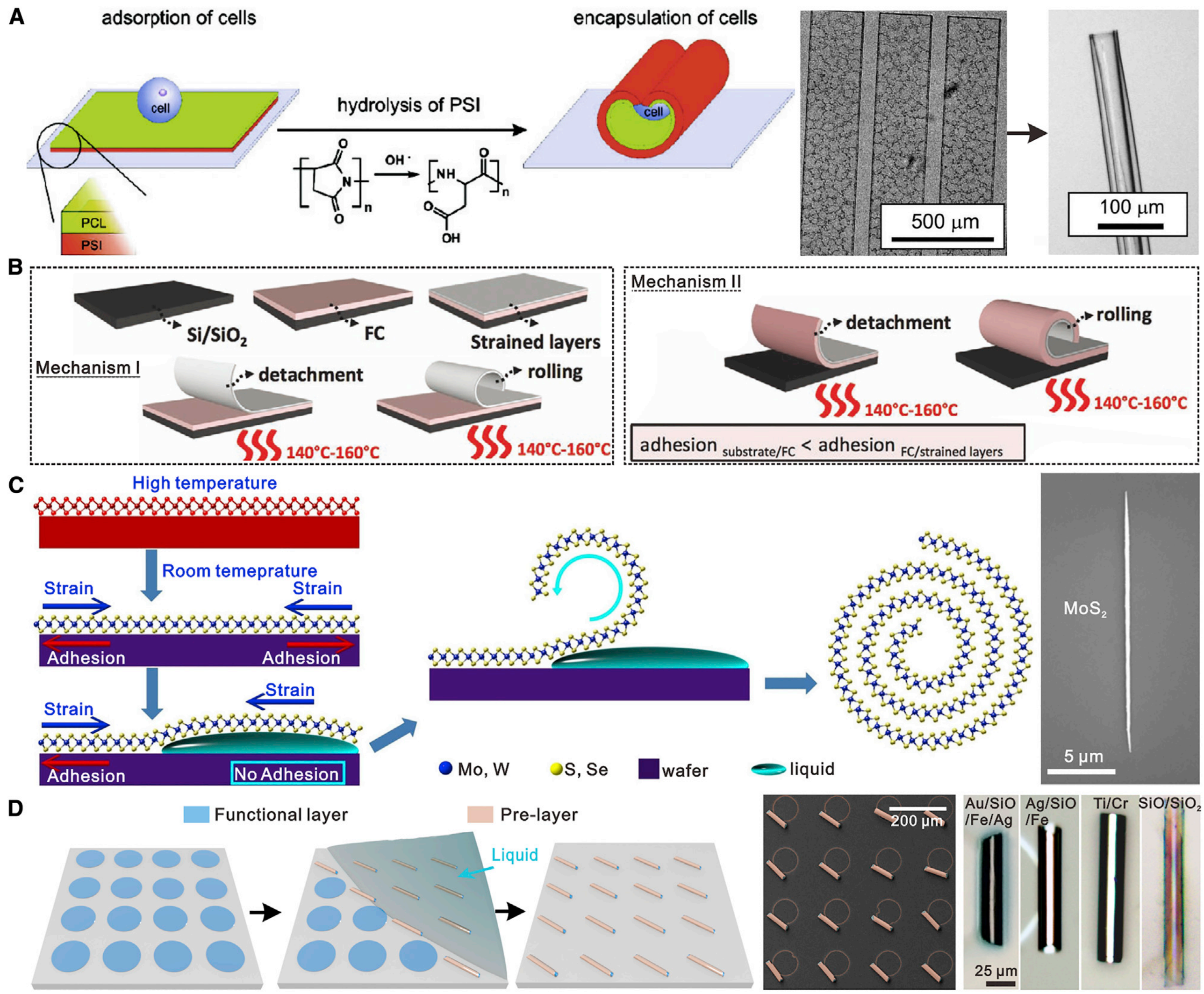

Figure 4. Rolling Origami without Sacrificial Layer

(A) Self-rolling of polymeric bilayer under the strain induced by hydrolysis in solution.[50] Copyright, 2011, American Chemical Society.

(B) Spontaneous detachment and rolling due to large thermal stress during heating. Mechanism I represents self-rolling of inorganic layer on expanded organic layer. Mechanism II represents self-rolling of upper layer together with organic layer.[51] Copyright, 2020, Wiley-VCH.

(C) Scrolling of TMD nanomembrane with insertion of liquid.[52] Copyright, 2018, The Author(s), CC BY 4.0.

(D) Spontaneous delamination and rolling due to liquid intercalation offering parallel and versatile rolling origami.[53] Copyright, 2019, The Author(s), CC BY 4.0.

upper layers. If the adhesion between substrate and FC layer is smaller, mechanism II happens and vice versa. Therefore, with careful selection of substrate and upper layers, 3D microstructures with different material combinations are obtained. This method excludes fluid to enter windings, effectively minimizing winding interfacial defects and enabling rolling origami of microtubes with multiwindings. The methods discussed above reflect that increasing strain gradient for rolling origami is commonly applied in the material systems containing organic layer, which has the ability to generate large additional strain to peel off upper nanomembranes.

To face the inorganic system, decreasing adhesion between nanomembrane and holding substrate is an optional strategy, which also can be achieved by environmental change. In these works, a tiny amount of liquid plays an important role as a

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lubricant between layers. Figure 4C illustrates that high-quality transition metal dichalcogenides (TMDs) nanotubes are fabricated with a nearly 100% yield by adding ethanol solution onto as-grown 2D TMDs.[52] Added ethanol solution intercalates into the TMD layer and substrate, releasing part of TMD layer. Owing to the ultrathin TMD layer, formed nanotubes have a diameter in tens of nanometer, which is much smaller than other reported rolled-up structures. And also, they achieve parallel production via patterning TMD with focused ion beam cutting and observe its outstanding optical and electrical properties benefiting from rolled-up nanostructures. Although the above example only shows rolling origami of TMD materials, the intercalation of liquid for delamination of a wide choice in material combinations is shown in Figure 4D.[53] Deposited multilayer nanomembranes are delaminated from the substrate, triggered by the intercalation of droplets. With proper selection of substrate, inorganic nanomembranes ranging from metals to oxides are successfully rolled up into 3D microstructures. These few two examples about decreasing adhesion by intercalation explain a simple and general method taking advantage of mild solution to roll a wide range of inorganic functional layers with little selectivity. The tiny amount of liquid used guarantees minimum damage on rolled-up micro-/nanostructures from surface tension. With a deeper understanding of the relationship between adhesion and internal strain, we believe this intercalation method will step further and become an important part of rolling origami.

Structure Designability

Origami is magical in turning 2D paper into desired 3D structures by hands. Different from a simple rolling manner, well manipulation in rolling process for diversified 3D structures is a key issue to realize rolling origami. It is well known that the size of rolled-up microstructures is altered by material characteristics as the thickness and internal strain, which is determined by deposition parameters.[54] But the other aspect, rolling direction, is not easy to control, as it is mainly defined in releasing process. And manipulation in rolling direction leads to vast types of rolled-up microstructures. Many explorations to well control rolling structures have been tried since the emergence of the first rolled-up nanotubes. We classify the rolling behavior control methods into two categories. One is manipulating nanomembranes before rolling, and the other is controlling rolling directions during releasing.

Nanomembrane-Based Control

In this part, we discuss the manipulation of nanomembranes toward directional rolling, including utilizing anisotropic nanomembranes and applying different pattern designs of isotropic nanomembrane. The anisotropy would determine final 3D microstructures because the preferring rolling direction of released nanomembrane is along the most compliant and energetically favorable direction. As shown in Figure 5A, deposited SiGe nanomembranes on n-Si (100) substrate are pre-shaped into strips whose long sides have different angles with the preferred rolling direction.[55] Directional rolling is due to anisotropic mechanical properties of single crystalline nanomembrane as well as an anisotropic etching of the sacrificial layer. Therefore, the middle strip released along the most compliant direction forms micro-/nanotube, although the other two form micro-/nano helical structures with the releasing direction deviating from the preferred rolling direction. In Figure 5B, rectangular-shaped InGaAs/GaAs bilayers are arranged in a wheel configuration, resulting in different types of rolling behavior due to the angle of pattern and preferring rolling direction.[56] Thus, patterning deposited nanomembranes according to prior rolling direction allows the fabrication of various rolled-up microstructures. On the other hand, it is mentioned that creating anisotropic nanomembrane requires high-quality deposition method, through which materials can be grown in single crystalline. This

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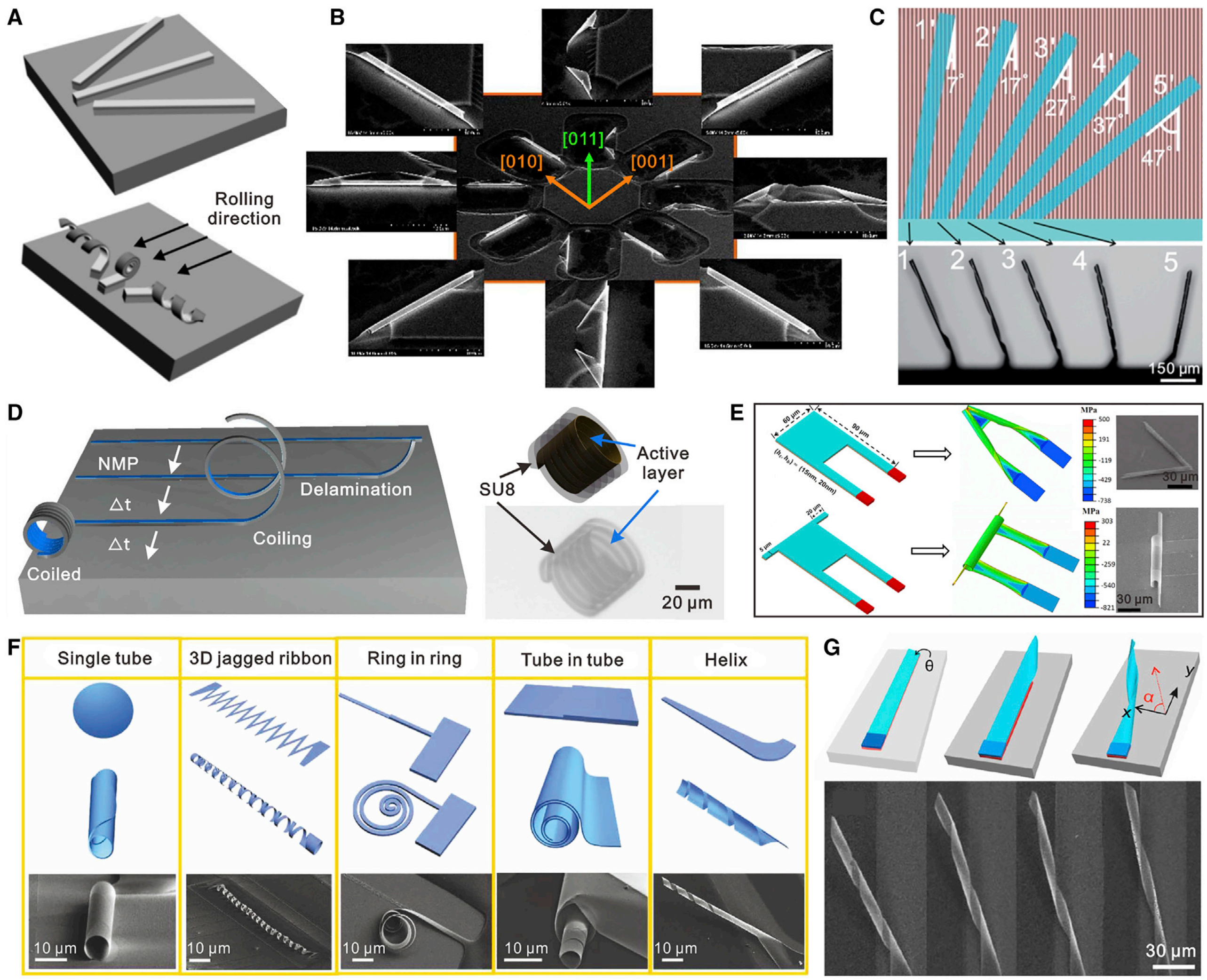

Figure 5. Structure Design under Nanomembrane-Based Control

(A) Formation of helical or tubular structure due to directional rolling.[55] Copyright, IOP Publishing.

(B) Directional rolling of monocrystalline nanomembrane with anisotropic properties.[56] Copyright, 2008, IEEE.

(C) Nanoimprinting nanomembrane with grating structure to guide rolling process.[57] Copyright, 2014, Royal Society of Chemistry.

(D) Fabrication of helical microstructure from long strip nanomembrane.[58] Copyright, 2011, American Physical Society.

(E) Corner attachment design to guarantee microtube formation.[59] Copyright, 2017, IOP Publishing.

(F) Flexible construction of rolled-up microstructure through different pattern design.[60] Copyright, 2017, Wiley-VCH.

(G) Precise control in rolling direction through wedge-shape pattern with tunable angle.[61] Copyright, 2018, American Chemical Society.

demand highly increases the cost of rolling origami and hampers this control method to be used in a blaze of materials.

Except for depositing nanomembranes with anisotropic mechanical properties, the anisotropy is also realized with post-modification. For example, wrinkled nanomembranes prefer rolling along the direction perpendicular to the long side of wrinkles, because rolling from the wrinkled edge needs an additional amount of energy.[62] The method to periodically wrinkle nanomembranes inspires the design of grating fine structures. Huang et al.[57] fabricated rectangular metallic nanomembranes and used a nanoimprinting technique to create grating structures on it (Figure 5C). The elastic energy barrier is higher when rolling the nanomembrane along grating lines compared to perpendicular to gratings. Therefore, grating structures determine a preferred rolling direction, resulting in designability in rolled-up microstructures. Through careful design of the anisotropy of functional layers, 3D geometry of tubular and helical microstructures could be controlled in a precise manner. Regardless of material systems compatible with anisotropic materials, the introduction of anisotropy may have some undesirable impact on their properties, leading to a recession in function.

Scientists also investigated controlled rolling from various 2D patterns of nanomembranes with isotropic elastic properties. As one of the most widely used patterns, rectangular shape is chosen as the beginning research object on the shape impact to 3D geometries. In 2005, it was found that the width, length, and thickness of an elastically isotropic nanomembrane determine whether a tubular structure or a coiled structure is formed.[63] As shown in Figure 5D, a compact helix coil is formed from a long (1 mm), narrow (7 μm), and thick (2–10 μm) strip.[58] Li et al.[45] further systematically illustrate the impact of geometry of a rectangular shape to the final 3D microstructures and find out that the width and length, together with rolling diameter, have a great influence on rolling behavior in a symmetric releasing system. Besides, Chen et al.[59] noticed a fixed rectangular-patterned nanomembrane starts rolling at its released corners, leading to a failure in tubular structure formation (upper in Figure 5E). They smartly designed an additional corner geometry to avoid this phenomenon. Comparing to a rectangular nanomembrane, nanomembrane with a bar at the corner achieves a higher unidirectional rolled-up yield, indicating the significant potential to optimize the yield of 3D rolled-up devices through the manipulation of 2D nanomembranes. Furthermore, a series of 3D structures were demonstrated by patterning isotropic nanomembranes with various shapes, as presented in Figure 5F.[60] This reflects that pattern design can effectively affect the final geometries of rolled-up microstructures. Then, a "utility knife" pattern is proposed to control the rolling direction of isotropic nanomembranes.[61] As illustrated in Figure 5G, a set of helices with various pitches is fabricated by changing the top angle of this pattern. This directional rolling is due to the anisotropic mechanical stresses generated with the contributions from the utility knife shapes.

It is concluded that to control rolling direction through deposited nanomembrane, in most cases, is to create strain/stress anisotropy. Breaking the symmetry of nanomembranes could not only be intrinsic by depositing single crystalline nanomembranes but also be extrinsic by introducing fine structures or patterning planar nanomembranes. What is more, the modification on nanomembranes, especially with interesting pattern design to form fantastic 3D microstructures, makes rolling origami more like an art in micro-/nanoworld rather than a fabrication technique.

Delamination Control

The key step of rolling origami is to release nanomembranes from the holding substrate. In a common situation, a sacrificial layer is set between deposited nanomembrane and substrate, which would be etched away in the releasing step. During the etching process, the corner/side that free first begin to roll advancing than other places. It is natural to consider that controlling etching anisotropy is an available route to control the rolling process. As an instance, etching a gap defined by photolithography at one edge of the pattern, named as etching window, creates an entrance for the etching solution to penetrate in, as shown in Figure 6A.[64] Then, the nanomembrane starts rolling from the etching window, realizing the manipulation of rolling direction. The same strategy is also achieved through glancing angle deposition (GLAD), as illustrated in Figure 6B.[32] Sacrificial layers are first patterned on the substrate, followed by the deposition of nanomembrane with glancing angle. Owing to the ballistic shadow effect from the patterned sacrificial layer, an etching window is created where no materials are deposited. Through this gap, etching solution

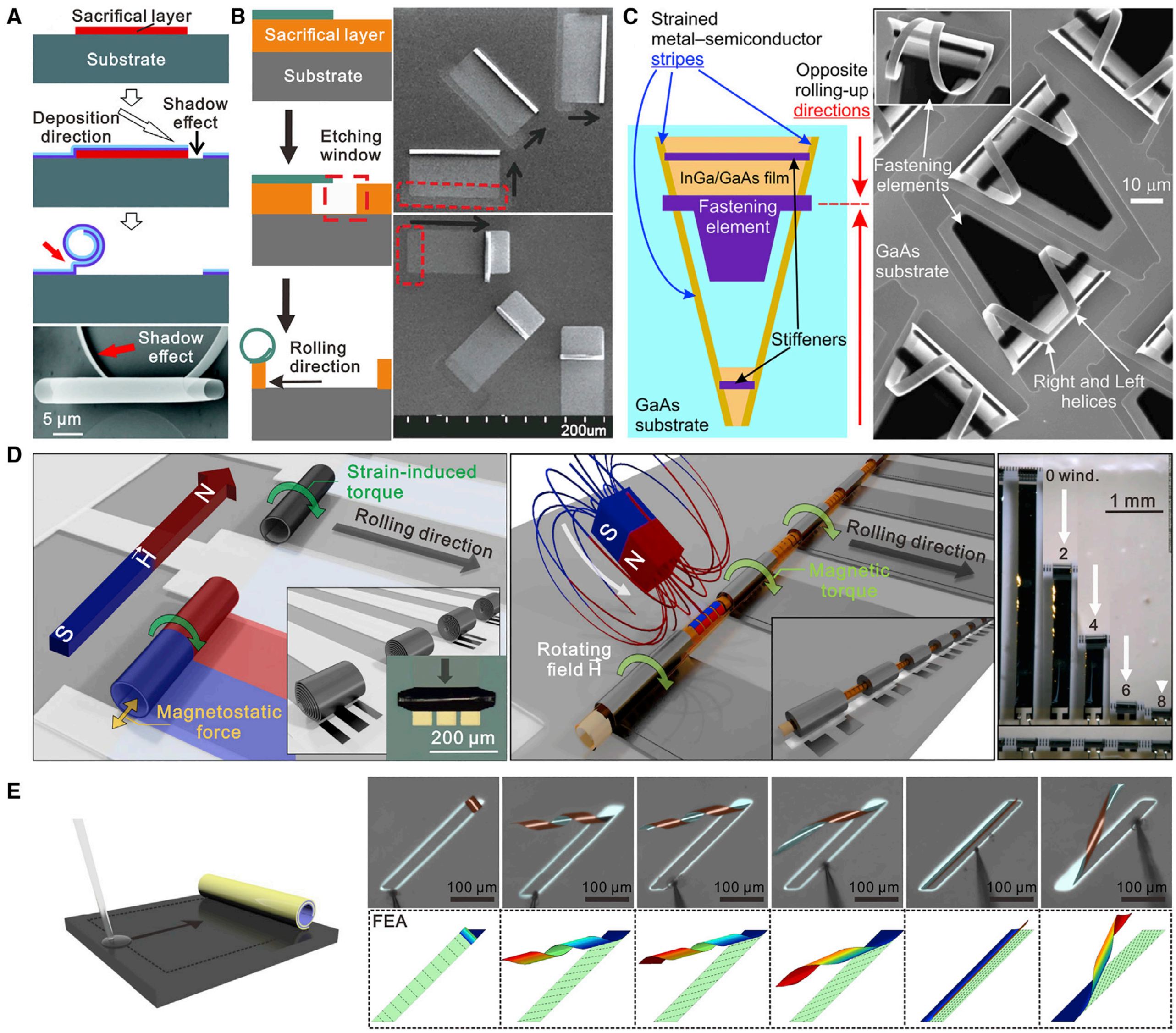

Figure 6. Structure Design under Delamination Control

(A) Creating etching window to guide the rolling of nanomembranes. Scanning electron microscope (SEM) images depict arbitrary rolling of square nanomembranes from etching windows (red dashed area).[64] Copyright, 2013, IOP Publishing.

(B) Directional etching starting from exposed window, created by shadow effect in glancing angle deposition.[32] Copyright, 2009, Royal Society of Chemistry.

(C) Additional fastening element design to force nanomembrane to roll from the other side.[65] Copyright, 2017, The Author(s), CC BY 4.0.

(D) Magnetic-guided rolling toward multiwinding tubes. Left panel depicts rolling process stabilized by an axially applied magnetic field. Right panel depicts rolling process totally manipulated by an external rotating magnetic field.[43] Copyright, 2019, The Author(s), CC BY 4.0.

(E) Precise control of rolling process by microdroplet-guided intercalation and delamination.[53] Copyright, 2019, The Author(s), CC BY 4.0.

penetrates sacrificial layer and encroaches the sacrificial layer from the side. In this way, upper layers start to delaminate from the gap side, realizing etching anisotropy. Compared with the previous method, GLAD combines material deposition with etching window creation, getting rid of the additional procedure to define the etching window.

As the effect of etching window is to accelerate the etching process in one direction, some methods are, on the contrary, suppressing rolling from the opposite direction.

Prinz et al.[65] creatively applied photolithography to design a fastening element, an element pressing on the nanomembranes like a stone on a paper, as illustrated in Figure 6C. It is noted that the fastening element is set near one side edge of the patterned nanomembranes. When the sacrificial layer is etched out from all directions in same etching rate, the rolling competition from two opposite sides is ceased by the fastening element. The side near the fastening element is stopped by rigid fixation, thus achieving unidirectional rolling. The fastening elements are also developed into many other structures. In another example, a fastening bar of photoresist at one side of deposited nanomembrane blocks the access of an etchant solution to the sacrificial layer.[66] In this way, etchant solutions are only allowed to start etching from the other side, thus causing the etching anisotropy. Except for assisting the rolling of nanomembrane in desired direction, the fastening element also has other functions. It keeps the 3D structures in position upon etching to construct stable free-standing 3D microstructures. These methods are proven to have the ability to fabricate highly ordered and uniform arrays, indicating a promising future for practical large-scale fabrication of 3D rolling origami.

If no etching control is applied, the final geometry would go toward misassembly, due to the degrees of freedom. This phenomenon leads to various pathways, especially for nanomembranes with high aspect ratio, making it difficult to generate an ideal tubular microstructure. Recently, external magnetic field is developed to restrain the degree of freedom during the rolling process and obtain high-performance microdevice, as presented in Figure 6D.[43] By applying an axial static magnetic field during assembly, ferromagnetic nanomembrane with high aspect ratio is prevented from an unwanted rolling shift in the axial direction by magnetostatic forces arisen in the magnetized windings. Besides, if radial magnetic field is applied, a defined nanomembrane attached to a pole-like magnetized rotor would achieve precise delamination, as seen in the right panel in Figure 6D. To be driven by the magnetic field, upper layers must involve a magnetic component instead of strain gradient for rolling. This method controls the rolling through directing magnetic functional nanomembranes in the whole delamination process, increasing overall assembly yield to a value larger than 90%. This kind of control is especially tailored for the rolling origami of nanomembranes with high aspect ratio to generate rolled-up microtubes with several windings.

With increasing popularity to roll nanomembranes without the assistance of the sacrificial layer, there is no necessity to control the etching process on these new fabrication methods of rolling origami. Instead, the delamination of nanomembranes, without the assistance of the sacrificial layer, can be controlled by manipulating the competition between strain and adhesive force. By introducing microdroplets to partially decrease the adhesion between deposited nanomembrane and substrate, precise control of delamination is obtained, resulting in smart construction of 3D structures with pre-designed geometries.[53] As presented in Figure 6E, the rolling direction is determined by the contacted point of microdroplet. The microdroplet intercalates from the contacted point and reduces local adhesion, triggering the rolling from the contacted point. This intelligent method, with the designability and predictability by quasi-static finite element modeling, achieves large-scale production of pre-designed 3D structures precisely, permitting a wide range of material combinations on different substrates. This kind of precise control during delamination takes one step forward to the ideal rolling origami.

Functional Diversity

After a full discussion about the fabrication technology of rolling origami, it is apparent that rolling origami permits a wide choice in materials with designable microstructure construction. Generally, resultant microstructures prepared from rolling origami include microtubes and helixes. These structures supply unique electrical and optical properties, along with property tunability due to curved behavior. Combined with different materials and their combinations, rolling origami achieves applications ranging from physical, chemical, to biological areas. For instance, metal applied in rolling origami serves as an important candidate in integrated circuits, magnetic devices, and tubular micromotors. Oxide material is essential in optics and allows direct recording of activities in a confined microenvironment. Semiconductor shows great potential in electron tunability, mainly applied in the device-required interaction between photon and electron. Besides, the realization of rolled-up 2D materials opens up a new era for tuning and sensing. Therefore, we plan to introduce these applications in material aspects to show the function diversity of the rolling origami system.

Metals

In planar integrated circuits, it is a conventional strategy to apply metallic layer for the construction of a simple electric connection and a bunch of passive or active on-chip electronic devices. Considering that rolling origami is a generic method to transform planar nanomembrane into 3D structures, metal-based, on-chip devices combined with rolling origami would provide enhanced integrated density due to reduced footprint area, together with outstanding performance.[67] In 2015, Karnaushenko et al.[68] proposed a 3D micro-antenna fabricated by self-rolling of a V-shaped metal strip. As shown in Figure 7A, the geometries of the antenna, such as diameter and winding numbers, are simply tuned by the optimization of strained bilayer and V-shape angle, which highly affects the performance of the antenna. They successfully implanted rolled-up micro-antenna with ca. 200 μm diameter in a tooth and achieved effective communication between implanted antenna with a smart phone. This demonstration highly reflects the unlimited potential of rolling origami in electrical devices, especially in those that require 3D configuration to function.

In these years, Li's group was focusing on the miniaturization of on-chip electric devices based on the circuits designed on the silicon nitride strained layer. Early in 2012, they developed a 3D inductor by rolling up Cu strip.[76] Rolled-up tubular structure provides a perfect spiral coil, endowing the device with good confinement of magnetic field and thus improved magnetic energy storage. Larger inductance was further obtained with optimal design and integration of ferrofluid magnetic materials through capillary force.[33] In Figure 7B, they reported another electrical microdevice as an inductor-capacitor (L-C) element fabricated by rolling origami.[69] This electrical microdevice is fabricated by rolling up a patterned metal strip into one tube. Perfect coupling in radio frequency is observed benefiting from confined 3D geometry. Hence, it is confirmed that rolling origami is regarded as an ideal platform to construct passive integrated components with reduced footprint area and enhanced properties.

Other than the elements discussed above, rolling origami with metals also performs excellent function in active devices as electrical sensors. Specifically, the hollow structure in rolled-up microtube establishes a well-defined microchannel for fluid sensing. In 2014, Martinez-Cisneros et al.[31] designed an impedance-based fluid microsensor by simply rolling up two metallic electrodes with oxides. It is noticed that, rather than simply detecting the impedance change due to fluid medium, this 3D microsensor performs available cell counting ability as the resistance changes with a single cell passing through. This makes it considered as a potential biosensor.[77]

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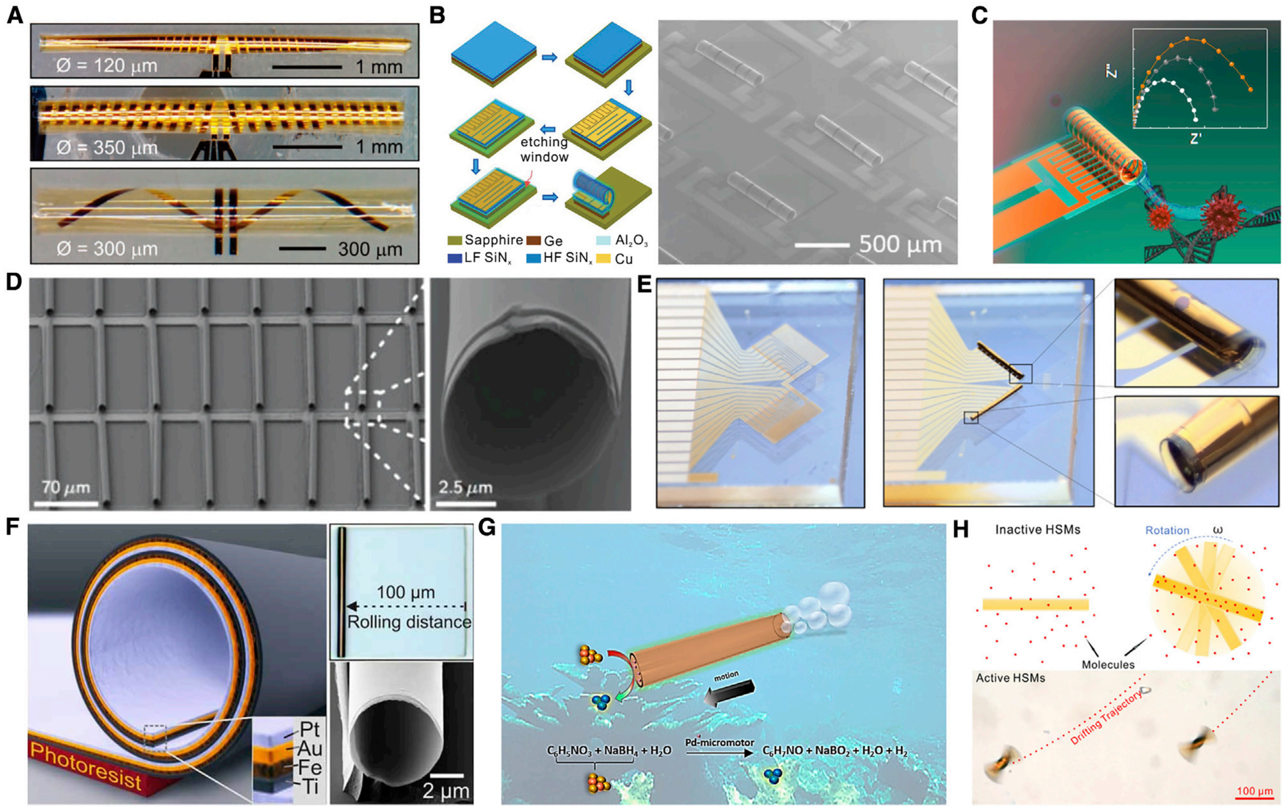

Figure 7. Rolling Origami Applications with Functional Metals

(A) Cu-wire-based, rolled-up microhelix as implant antenna.[68] Copyright, 2015, Springer Nature.

(B) 3D radio-frequency L-C circuit with ultra-compact footprint.[69] Copyright, 2020, Wiley-VCH.

(C) Rolled-up circuits for DNA detection through electrochemical impedance spectroscopy.[70] Copyright, 2016, American Chemical Society.

(D) Rolled-up microtubes consisting of Ni and permalloy.[71] Copyright, 2013, Wiley-VCH.

(E) Magnetic microtubes as GMR sensors for angular encoders.[72] The Author(s), CC BY 4.0.

(F) Rolled-up tube with Pt catalyst for micromotor.[73] Copyright, 2009, Wiley-VCH.

(G) Water decontamination by rolled-up micromotor with functional Pd.[74] Copyright, 2015, American Chemical Society.

(H) Motile micromotors for ultrasensitive SERS sensing.[75] Copyright, 2020, American Chemical Society.

Moreover, with further biomodification on Au surface, similar rolled-up circuit integrated into a microfluidic chip is able to detect DNA content with a chemical impedance spectroscope (Figure 7C).[70] In this report, they successfully detect H1N1 virus DNA in ultralow concentration through this impedimetric DNA sensor and would extend to other types of DNA sensing with proper modification. Therefore, combined with conventional planar circuit design and functional modification, shape transformation via rolling origami offers a simple and compatible route to build all kinds of integrated 3D circuits and functional components.

Another intriguing property of metal is the ferromagnetic properties brought from some elements as Fe, Co, and Ni. Rolled-up ferromagnetic microtubes in Figure 7D present an unconventional magnetic performance due to the additional stress introduced by curved geometry.[71] Further applications were carried out based on the giant magnetoresistive (GMR) effect in ferromagnetic materials. With simply attaching two electrodes on ferromagnetic nanomembranes, a rolled-up tube is able to detect ultrasmall magnetic field recognized by resistance variation. And the integration of two rolled-up microtubes orientated different directions (Figure 7E) enables 3D magnetic field vector sensing with ultrahigh sensitivity. And with a deep investigation, it is believed that ferromagnetic microstructure from rolling origami will create stupendous opportunities in magnetic-related areas.

In microworld, efficient and controllable motion of artificial micro-object is a fascinating task to realize advancing applications at such a scale. Catalytic reaction based on noble metals offers an opportunity to generate sufficient propulsion power by utilizing surrounding medium.[78] In 2009, Solovev et al.[73] successfully achieved direction motion of a rolled-up microtube in a liquid environment (Figure 7F), which is considered as the first example of micromotor established on bubble propulsion mechanism and paves the way to highly efficient micromotor with ultrafast speed. And the addition of ferromagnetic layer offers motion control ability through external magnetic field. Such tubular micromotor is decorated with Pt layer or nanoparticles in the inner wall, which decompose H_2O_2 in surrounding environment into oxygen. Benefiting from confined cylinder microchamber, oxygen gathers together inside the tube and produces bubbles in high efficiency. Then, continuous ejection of generated bubbles leads to valid locomotion of micromotor.

As for the application of micromotors, scientists mainly focus on their great potential in environmental and biomedical areas by modifying tubular walls of micromotor with different functional materials. Figure 7G presents an environmental application in which micromotors can decompose nitroaromatic pollutants with Pd catalyst inside the tube.[74] Similarly, applying Fe in micromotor system offers the ability to degrade organic pollutants by Fenton reaction.[79] These experiments all reflect improved efficiency in water decontamination compared with static particles, which is attributed to the automatically collecting effect during moving. Considering this effect, an ultrasensitive surface-enhanced Raman scattering (SERS)-sensing application with tubular micromotor is proposed (Figure 7H).[75,80] After self-propelled moving and collecting molecules for a period of time, collected target on the outer wall of microtube is detected by Raman spectrum. Signal of molecules is largely enhanced due to enrichment manner and SERS from surface-modified Au layer. Such micromotors can be applied in environmental monitoring. Moreover, tubular micromotor is designed as a nanotool for bioapplications.[81] Xi et al.[82] obtained a microdriller by rolling up a magnetic micromotor. The micromotor drills into organ by applying a rotational magnetic field to meet the requirement of minimally invasive surgery. The combination of rolled-up microstructures with cells as sperm provides another solution for biocompatibility in biomedical applications and is expected to be applied in revolutionized assisted-reproduction technology and nanomedicine.[83,84]

Oxides

Unlike metals, oxides often possess a transparent appearance, making them a good candidate in optics. Benefiting from the uniform distribution of strain in a layered system, nanomembrane after rolling origami prefers to transform into an ideal cylinder shape with negligible asymmetric geometry at the edge. Thus, rolled-up microstructure is considered as an optical cavity with whispering gallery modes. A rolled-up microcavity system commonly consists of SiO_x or TiO_2 nanomembrane for rolling, sometimes together with a further coating of Al_2O_3 or HfO_2 to enhance optical properties. The subwavelength thickness of the tubular wall indicates a strong coupling between the optical field inside tube and external environment through evanescent wave. And several optical sensing applications were proposed to detect surrounding signals as fluid properties and humidity.[85] Additionally, molecules attached to the tubular wall can be coupled into a non-evanescent region, leading to remarkable

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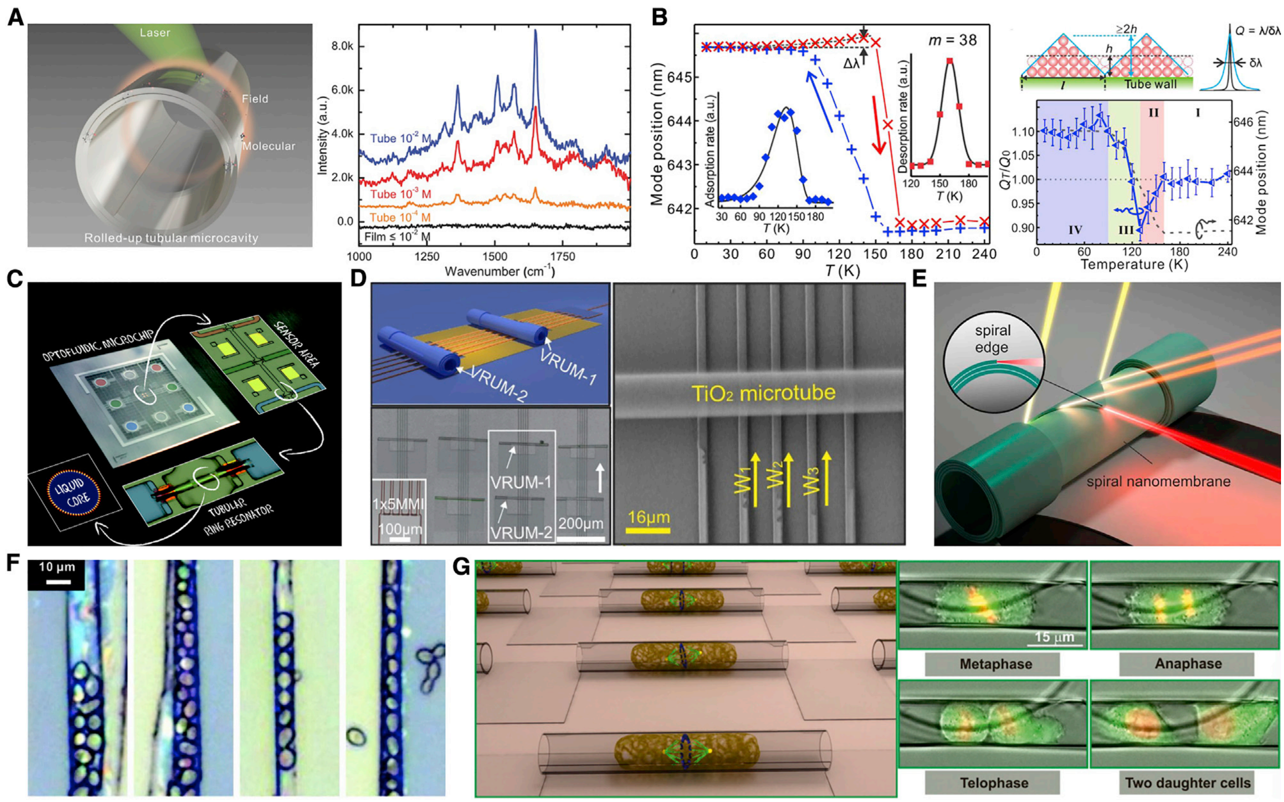

Figure 8. Rolling Origami Applications with Transparent Oxides

(A) Enhanced Raman sensing due to resonance coupling from oxide microcavity. Raman spectra depict the enhancement by tubular structure.[86] Copyright, 2016, Wiley-VCH.

(B) Optical sensing by rolled-up microcavity to expose water nanostructure formation. Left panel depicts resonance mode shift during water growth and desorption triggered by temperature change. Right panel depicts four regions of water layer formation revealed by resonance variation.[87] The Author(s), CC BY 4.0.

(C) Integrated on-chip optofluidic sensor with rolled-up microtube.[88] Copyright, 2012, Royal Society of Chemistry.

(D) Monolithic integration of rolled-up microcavity on a photonic chip.[89] Copyright, 2018, Optical Society of America.

(E) Directional light emission from spiral microcavity.[90] Copyright, 2019, American Chemical Society.

(F) Transparent microtubes as confined scaffold of yeast cells.[64] Copyright, 2009, Royal Society of Chemistry.

(G) Single-cell study in functionalized rolled-up microtube.[91] Copyright, 2014, American Chemical Society.

enhancement in its optical signals (Figure 8A).[86] And in the opposite, the intrinsic resonance of rolled-up microtube is highly influenced by attached matters, bringing about inventions of dynamic molecule sensing on the surface.[92] Recently, Yin et al.[87] investigated water nanostructure formation on amorphous oxide surface by analyzing peak shift and quality factor (Q factor) during water sorption and desorption. In the left panel of Figure 8B, they found that peak position varies during temperature rising and decreasing, which is attributed to water sorption and desorption. Thus, the region of temperature with a water dynamic process is confirmed. Analysis in Q factor variation (right panel in Figure 8B) further reflects a detailed sorption process, in which a decrease of Q factor refers to the generation of nanocluster on the surface, and later increase refers to the formation of a smooth water layer. With this optical measurement, molecule dynamic process is fully revealed, elucidating that microcavity fabricated by rolling origami is a powerful tool in sensing area.

With these outstanding sensing properties, it is envisioned that the integration of rolled-up microcavity into an on-chip system will lead to promising results in optical systems. In 2012, Harazim et al.[88] tried to transfer single rolled-up microcavity into a microfluidic chip (Figure 8C). Benefiting from the hollow structure, the transferred microtube serves as an individual cavity to detect refractive index change in the surrounding environment and, in the meantime, as a microchannel to allow microfluid flowing through. As fluid going through the rolled-up microtube, resonance mode change directly reflects the properties of fluid, without requirement of further labeling or other treatment. Furthermore, a monolithic integration in a photonic chip was reported in 2018,[89] as shown in Figure 8D. Instead of transferring a prepared microtube, the authors applied rolling origami to directly transform deposited nanomembranes on photonic chip into rolled-up microcavities, coupled with planar photonic waveguides beneath them. Such an integrated photonic chip presents high sensitivity to liquid medium, as well as tunability in optical modes.

Except for investigation for practical applications, some unique optical phenomena are also observed in the rolled-up microcavity. Unlike other cavities, rolling origami creates natural asymmetric geometry in whispering gallery mode (WGM) resonance, owing to the rolling edge as mentioned above. Fang et al.[93,94] theoretically found that rolling edge at inner and outer walls highly influence light propagation behavior in clockwise and counterclockwise manners. Thus, there exists a coupling between overlapping area and optical resonance,[93] accompanying with exceptional points due to asymmetry.[94] And also, this rolling edge breaks perfect resonance in the tube wall, leading to directional light emission at the edge (Figure 8E).[90] The authors designed a triangle shape to alter overlapping distance, through which light could emit in different directions at different edge points. A similar concept was applied to create additional confinement along the tubular axis by a lobe design,[95] thus endowing rolled-up microcavity with 3D light confinement ability. With improved fabrication methods and clever design toward high-quality rolled-up oxide microcavities, enhanced optical properties will produce a bunch of applications spanning from advanced environmental sensing to novel optical research.

Oxide-based rolling origami also provides an ideal transparent microchamber to directly observe the evolution of cells, especially to investigate cell growth in a confined environment. As early as 2009, Huang et al.[64] observed the growth of yeast cells in SiO/SiO_2 microtube (Figure 8F). They found that, in different confined conditions as tunable tube diameter, yeast cells exhibit different growth behavior. A deeper cell study was carried out by Xi et al.,[91] as shown in Figure 8G. With post-modification of rolled-up microtubes, they created a proper environment for cell culture. With a high-resolution microscope, the division of mammalian cells was recorded in detail, and they found that spatial confinement has an effect on the fidelity of chromosome segregation. And in these years, different types of cells were studied in such transparent rolled-up microtubes,[96,97] illustrating that microstructures fabricated by rolling origami can serve as a reliable cell scaffold in the biological area.

In addition, there are some energy storage applications based on the oxide rolling origami system.[98] For example, applying oxide nanomembrane to construct a dielectric capacitor and transforming it into rolled-up microtubes would largely reduce the size of on-chip device, as well as provide enhanced capacitance due to overlapping turning.[99] Besides, rolled-up oxide nanomembranes are an optional choice as the anode and cathode in lithium ion battery (LIB)/sodium ion battery (SIB) systems due to its high specific surface area and tolerance to the extreme volume change during charging and discharging.[100] It is found that rolled-up nanomembranes consisting of SiO_x provide excellent stability serving as an anode in

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LIB.[101] And cathodes made of rolled-up MnO_x nanomembranes are proven to improve the capacity and stability of LIB.[102] These advantages are also brought to other material systems for appropriate subjects.

Semiconductors

Semiconductors, regarded as the fundamental and functional units in electronics nowadays, have indispensable band structures and carrier mobilities. And related nanostructures generate a set of new physical properties. When semiconductor materials meet rolling origami, unique properties emerge, leading to various applications in 3D types. The first one is the strain engineering due to curved nanomembrane. If a flat nanomembrane rolls into a cylinder structure, strain inside will be redistributed, leading to lattice constant change that affects band structure and other properties.[103] In Figure 9A, strain effect on the conduction band of quantum well is illustrated.[104] Apparently, the quantum well conduction-band edge is tilted because of rolling behavior, leading to a redshift in photocurrent response spectra. This result reflects that rolling origami has the ability to modify the physical properties of semiconductor with strain engineering. And the scale of strain can be simply manipulated by radius control in fabrication process. In another research, abnormal magnetotransport in rolled-up 3D microstructure is reported.[105] 2D electron gas with high mobility is generated in rolled-up microhelix with quantum wells. It is found that a strong asymmetric longitudinal magnetoresistance appears in this microhelix, as presented in the inset of Figure 9B. Therefore, applying rolling origami into semiconductor materials can produce tantalizing properties with 3D hollow structure and curved surface.

Interaction between light and electron in semiconductor materials is another important aspect in optoelectronics. With quantum well or quantum dot design, spontaneous light emission is observed with optical or electrical excitation. As discussed in Oxides section, tubular microstructure fabricated by rolling origami provides a WGM resonator to highly confine light propagation. Thus, light emission efficiency is improved with a combination of quantum well/dot and rolling origami in semiconductor system (Figure 9C).[112] Furthermore, emission properties can be manipulated by strain engineering, which is determined by geometric parameters,[113] or coupled with other optical material systems, like hyperbolic metamaterials.[106] Based on the enhanced emission in rolled-up WGM resonator, Mi's group developed several semiconductor tube laser prototypes pumped by light[114] or electricity[107] (Figure 9D) with an ultralow threshold.

Other than photon generation from electrons, the opposite conversion is also vital in the applications as photodetectors. In 2016, Wang et al.[108] proposed a 3D infrared photodetector by rolling up a quantum-well-embedded nanomembrane (Figure 9E). Compared with planar photodetectors, tubular photodetector presents an improved responsivity and detectivity owing to light confinement. And it is noticed that this tubular geometry allows a wide incident angle detection, realizing spatial detection of light that is hardly achieved by 2D device. These works enlighten us that transforming planar nanomembrane devices into 3D structures through rolling origami not only improves their original performance but also provides novel abilities as well as a new degree for device design.

2D Materials

In recent years, the development of 2D materials induces an innovation in material and engineering areas. Realizing that its ultrathin planar structure is similar to nanomembranes, 2D material is a considerable candidate in the rolling origami system.

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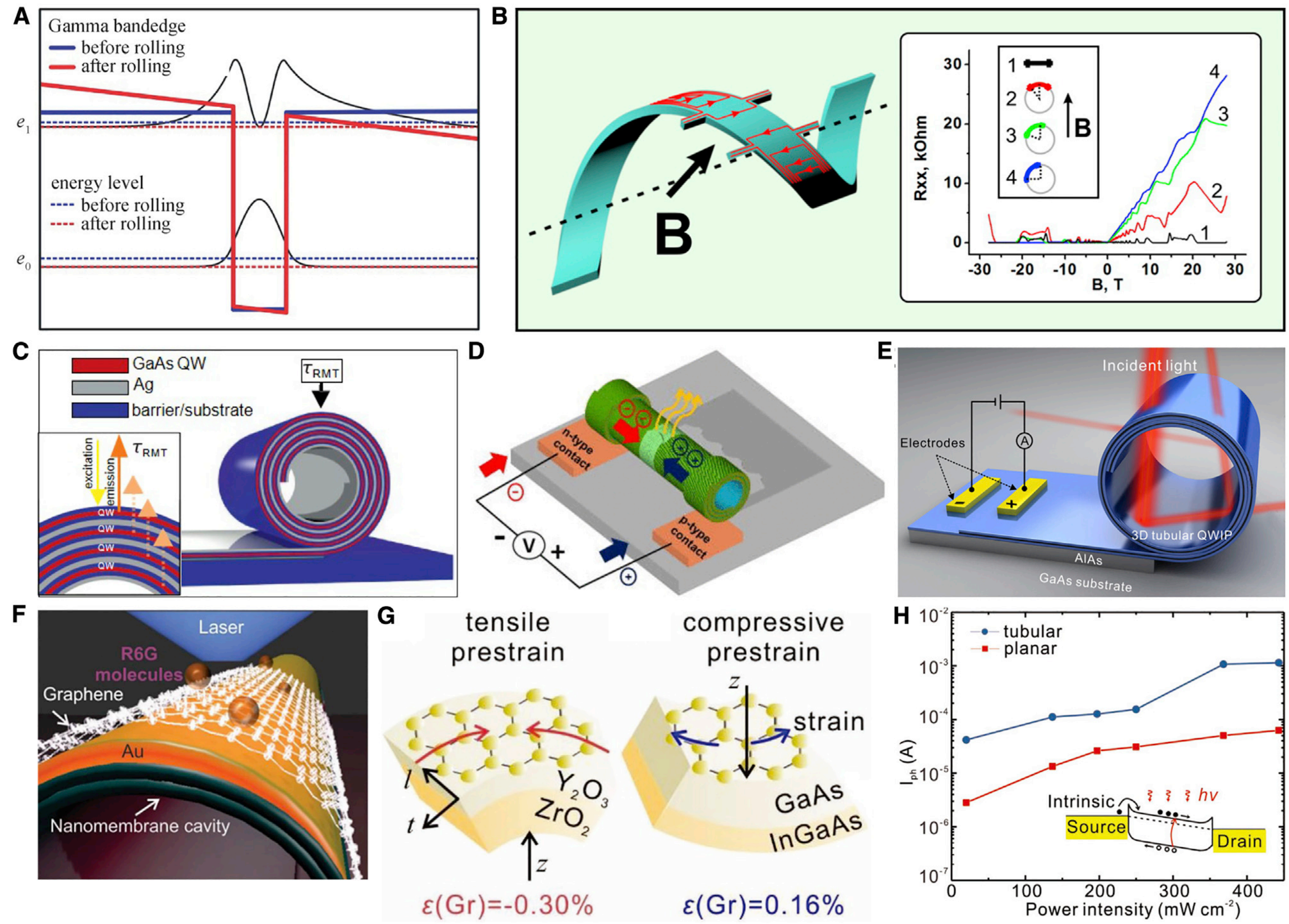

Figure 9. Rolling Origami Applications with Semiconductors and 2D Materials

(A) Conduction-band diagram illustrating tilted quantum well band edge due to strain redistribution in rolled-up microstructure.[104] Copyright, 2017, Chinese Institute of Electronics.

(B) 2D electron gas in rolled-up microhelix. The inset depicts different magnetoresistance of 2D electron gas in microhelices with different rotation angles.[105] Copyright, 2015, American Chemical Society.

(C) Spontaneous light emission in rolled-up hyperbolic metamaterials with quantum well.[106] Copyright, 2016, American Physical Society.

(D) Microtubular laser generated by electrical injection.[107] Copyright, 2015, AIP Publishing.

(E) 3D wide-angle infrared photodetector achieved by rolled-up quantum-well-embedded nanomembrane.[108] Copyright, 2016, The Author(s), CC BY 4.0.

(F) Optoplasmonic microcavity decorated with graphene for organic molecule detection.[109] Copyright, 2019, American Chemical Society.

(G) Enhanced $MoSe_2$ tubular photodetector. Inset illustrates the band diagram of $MoSe_2$ photodetector device.[110] Copyright, 2019, Wiley-VCH.

(H) Arbitrary strain tuning in graphene through different rolling system design.[111] Copyright, 2019, Wiley-VCH

As discussed in the Material Compatibility section, rolled-up 2D materials as TMD can be simply fabricated through the liquid intercalation method and perform improved optoelectrical properties.[52] The next step is to integrate them into on-chip electronics for potential applications. Yin et al.[109] transferred graphene onto a rolled-up nanomembrane to construct an optical sensor (Figure 9F). Rolled-up tubular structure with Au surface coupled with graphene constitutes a hybrid opto-plasmonic cavity, in which electric field is significantly enhanced. The enhancement leads to molecule-level detection of organics by optical signal, and they demonstrated dynamic monitoring of photodegradation progress on the tube surface.

Furthermore, rolling origami also has the ability to force 2D materials rolled up *in situ* to directly assemble 3D microdevice. Zhou et al.[110] designed a $MoSe_2$ rolled-up microtube and applied it as a photodetector. Two parallel metal strips are designed and deposited on $MoSe_2$. This nanomembrane system is released with selective etching as $MoSe_2$ was grown on Ge substrate. Deposited metal strips not only provide additional strain gradient for rolling but also serve as electrodes to output optoelectrical signal. Figure 9G reflects that the $MoSe_2$ layer in tubular geometry performs higher photocurrent compared with the planar one, which is on account of enhanced optical absorption and higher surface states density. Besides, as shown in the inset of Figure 9G, electronic band structure modification due to the rolling process is illustrated.

Strain engineering in 2D material is one of the most attractive issues to alter its properties for optimal performance. In previous research, 2D material is commonly transferred onto a flexible polymer substrate and is stretched or bent through mechanical deformation of the polymer substrate. Although the properties of 2D materials under different strain states can be fully investigated via these experiments, these stressed 2D materials are hardly compatible with on-chip integration, let alone practical applications. Wang et al.[111] overcome this drawback by fabricating rolled-up graphene microtube through rolling origami. The strain state in graphene is successfully altered from compressive to tensile strain, with proper design of deposited nanomembrane system to force graphene rolled up or bent down (Figure 9H). Hence, Raman signal of graphene varies with the strain state change.

Summary of Functionalization of Rolling Origami

In this section, we summarize functionalization of rolling origami among several material sorts and find out all kinds of materials have their potentials in different areas. And in this section, we want to give a brief discussion about which areas are suitable for rolling origami. First is on-chip microdevices, including electronic, magnetic, and optical components. Comparable fabrication process in rolling origami makes this technology very suitable to construct on-chip electronic microdevices. The transformation from 2D to 3D offers designability with an additional dimension and also allows conventional complex planar patterns. Thus, conventional passive and active devices consisting of metals,[15,68–70] semiconductors,[108,115,116] and 2D materials[109,110] are reorganized into 3D types with reduced area and improved performance. Energy storage applications, including supercapacitors and LIB/SIB, are also developed using oxides,[99,101,102,117,118] silicon,[119,120] and other materials.[19,121] On-chip magnetic microdevices constructed from rolling origami mainly focus on GMR sensor.[21,41,72] In the optical area, cylinder geometry in rolling origami induces good light confinement through WGM resonance with potential in light emission.[90,107,] Impressive optical sensing applications with sensitivity up to monomolecular level are demonstrated, which have the capability of integration on a photonic chip.[87,89,92,122,123] As for bioapplications, transparent microtube is regarded as a reliable chamber and scaffold for single-cell study.[64,91,96,124] Another important area is off-chip micromotors with self-propelled locomotion. Micromotors are used for environmental areas as pollutant monitoring and delamination.[74,75,79,80] And these biocompatible micromotors are developed as *in vivo* nanotools for drug delivery and microsurgery.[82–84] The investigation on 2D-material-based rolling origami is currently preliminary. But it is predictable that rolled-up 2D material device has infinite potential in optics and electronics, taking advantage of strain tunability and on-chip integration of rolling origami.

Toward Smart Systems

What is the biggest difference between cylinder microstructure from self-assembly method and rolling origami? It seems that both methods can produce microtubes

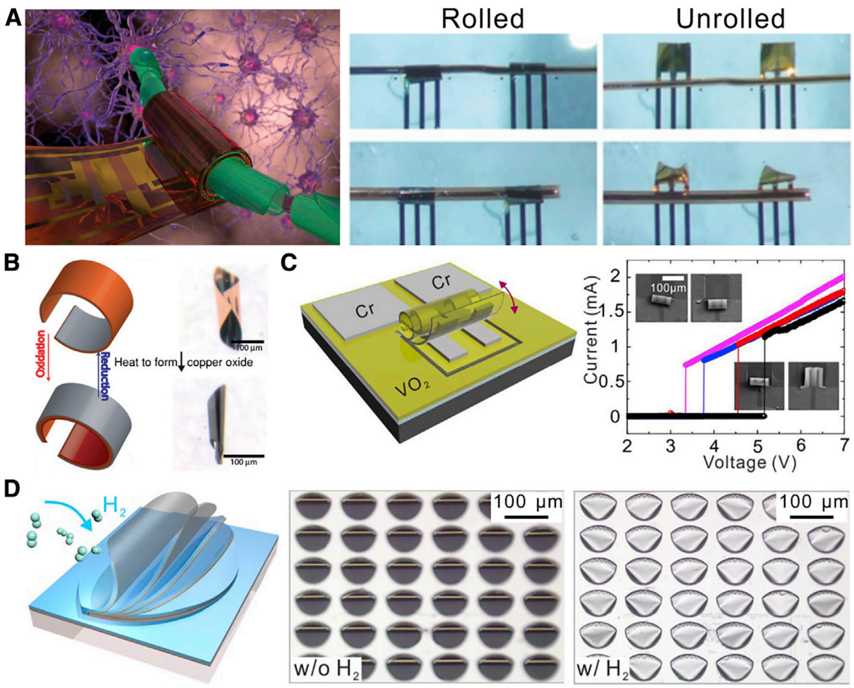

Figure 10. Rolling Origami toward Smart Systems and Applications

(A) Hydrogel-based rolled-up microtubes with reconfigurable properties due to swelling effect. Rolling behavior is controlled by adjusting the content of isopropanol in water solution.[42] Copyright, 2015, Wiley-VCH.

(B) Active shape transformation due to oxidation and reduction reaction of Cu.[127] Copyright, 2010, Wiley-VCH.

(C) Reconfigurable nanomembrane established on the phase transition of VO_2. Right panel depicts phase transition property of VO_2 nanomembrane and corresponding shape transformation.[128] Copyright, 2018, American Chemical Society.

(D) Spontaneous shape changing in hydrogen environment due to volume expansion of Pd.[129] The Author(s), CC BY 4.0.

and microhelices in all kinds of materials. Once you realize that rolling origami is a strain-dependent manner, it is revealed that rolling origami microstructure is sensitive to the strain distribution in nanomembrane system. As summarized above, previous reports mainly focus on the exploration of the properties and functions of rolled-up microstructures and try to optimize the size and geometry of structure through fabrication parameters although are rarely aware of dynamic shape changing in rolled-up microstructures.

Smart material is a type of material of which properties change with specific stimuli, especially mechanical properties. The volume or lattice change of smart material with stimuli can generate effective deformation, which is widely applied in microactuators and robots. Rolled-up microstructure is endowed with shape reconfiguration and tunability if rolling origami is combined with smart materials.[125] Hydrogel is a well-known smart material due to its swelling ability with volume expansion. In 2015, Makarov et al.[42] incorporated active hydrogel into rolled-up circuits to achieve a reconfigurable 3D implant device (Figure 10A). Its diameter and rolling states are tuned by environmental conditions as solution composition and pH, owing to different swelling behaviors. With diameter tunability and biocompatibility, hydrogel-based rolled-up microtubes are adaptive to enclose and guide the growth of nervous fibers with size ranging from 10 to 50 µm. And integrated functional and logic circuits in rolled-up microtubes are proven as a powerful tool to detect ionic signals, which is envisioned in the diagnostic of neuronal activity. Moreover, based on diverse swelling behavior in the hydrogel family, the geometry of rolled-up microstructures can be altered by all kinds of signals, which would largely expand their application scenarios.[126]

As for inorganic materials, several mechanisms were proposed to trigger deformation in nanomembrane system. As shown in Figure 10B, Gracias et al.[127] designed a chemically triggered rolling transformation due to surface oxidation and reduction reaction. They built Cr and Cu bilayer system. As the nanomembrane heated in oxygen atmosphere, Cu is oxidized into Cu_2O with the generation of external strain, leading to the reverse bending of nanomembrane. When moved into hydrogen atmosphere, the material was reduced to Cu again and the nanomembrane returned to the original state. Combined with pattern design with photolithography, further complex folding structures were obtained with localized bending.

Furthermore, inorganic materials with phase transition properties were applied in rolling origami. These materials provide ultrafast stimuli-responsive ability as phase transition is much faster than diffusion process in swelling. And higher rigidity of inorganic materials makes them superior in miniaturized self-supported 3D structure. Tian et al.[128] developed on-chip inorganic microactuators with vanadium dioxide (VO_2), which has metal-to-insulator transition by heating (Figure 10C). With a simple circuit design in rolling origami fabrication, this VO_2-based microactuator is controlled by heating or electrical signal trigger. Besides, they noticed that triggered temperature or required input energy decreases with smaller diameter of fabricated rolled-up nanomembranes, which is attributed to strain-tuning ability in phase transition properties. Xu et al.[129] found that the volume expansion of palladium (Pd) in hydrogen environment can be integrated into rolling origami. As schematic illustrated in Figure 10D, rolled-up nanomembrane incorporated with a Pd layer varies from rolled-up state to planar state with hydrogen injection. The unified responsive behavior of arrayed nanomembranes is observed, which leads to an obvious change in transmittance and difference observed by naked eyes. Benefiting from the rapid transition process, they developed a rapid and sensitive hydrogen detector without any energy consumption.

A typical 3D structure fabrication method named as 3D printing has been widely applied in numerous areas ranging from macroscale to nanoscale, through which assigned materials are directly printed on a platform to construct complex 3D structures.[130] And introduction of smart materials into 3D printing brings about talented technology that shapes vary with time, known as 4D printing.[131] Similarly, it can be seen that the combination between rolling origami and smart materials brings additional time degree to rolled-up structures. Nowadays, smart materials applied in rolling origami cover a bunch of catalogs. As we discussed above, rolled-up microstructures with smart hydrogels can change their shapes with swelling behavior triggered by different signals as pH and temperature. And using chemical reaction to change the properties of partial materials also leads to the shape transformation. Besides, the phase transition properties of some specific materials as VO_2 are considered as an outstanding option for its ultrafast speed and large deformation. With this dynamic ability of rolled-up structures, various applications from micromechanical to environmental and biological areas were realized and are expected to be applied in robotics. Except for taking advantage of these large deformations in the rolling origami system, using optical or electrical methods to recognize small deflection in structures will bring about a new type of sensors with incredible performance.

Summary and Outlook

With 2 decades of development in rolling origami, this family is filled with all kinds of materials and structures and derives a bunch of inspiring applications. The key in fabrication is peeling off strained nanomembrane from substrate. This peeling off process can be achieved by selectively etching underneath sacrificial layer. So, to get rolled-up microstructures in different materials, the choice of sacrificial layer must be carefully concerned. Spontaneous delamination without sacrificial layer is an optional choice, although deep consideration in the competition between adhesion and strain distribution is required. In addition, applying 2D pattern design with rolling direction control, a flat nanomembrane can be rolled into various interesting microstructures. Pattern design can be realized by simply covering a mask or applying standard photolithographic procedures. And rolling direction control needs to be achieved by altering the properties of nanomembrane itself or adhesion ability to substrate.

On the base of versatility in materials and structures, it becomes natural to develop numerous functions and applications spanning from electronics and optics to mechanics and biology. Two advantages of rolling origami need to be highlighted. One is that rolling origami is an *in situ* method to transform fabricated planar nanomembranes or devices into stereo types, getting rid of other transferring processes. The others are the unique physical properties in spiral microstructures as confined boundaries from tube for light propagation and confined geometries for chemical reaction and cell growth. Also, strain engineering due to rolling origami leads to exciting results in semiconductor and 2D materials.

Still, the versatility of state-of-the-art rolling origami is not enough to meet the demand for "real" rolling origami. A real rolling origami fabrication should be a convenient method with considerable cost to fabricate desired rolled-up microstructures with arbitrary designability in materials and structures. Fabrication methods based on inorganic or organic sacrificial layers have been explored for years with various structures and applications, although the issue stands there that rolling direction is hardly controlled in an ideal manner as etching process is commonly based on the diffusion of liquid or gas. On the other hand, direct delamination methods show their potential in arbitrary rolling origami, but further study is highly required to improve its capability toward complex rolling structures as integrated circuits. The development in the fabrication process of rolling origami would bring about novel structures with unpredictable properties. Moreover, combined with smart materials, structure constructed from rolling origami becomes an interdisciplinary platform. Electrical and optical properties revealed before, together with biological applications, are now linked to the dynamic deformation of structure. Beyond establishing a lab-in-a-tube system, building a smart system in rolled-up microstructures will be an indispensable part of future 3D microstructure-based applications.

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AUTHOR CONTRIBUTIONS

B.X. and X.L. contributed equally. B.X. and Y.M. conceived the idea. B.X., X.L., and Y.M. wrote the manuscript and prepared the figures.

DECLARATION OF INTERESTS

The authors declare no competing interests.

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