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Strain-engineered stretchable substrates for free-form display applications

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Abstract

With the growing potential of the Internet of Things, displays are being utilized to provide various types of information in every aspect of daily life, leading to the expansion of form-factor-free displays. Stretchable displays are considered the ultimate goal in form factor innovation, and they are not limited to rectangular shapes with deformation characteristics suited to target applications. Because reliable stretchable displays should be robust under uniaxial and biaxial strain, there have been efforts to tailor mechanical stress with promising strategies from structural and material perspectives. This review focuses on strain-engineering stretchable substrates for free-form display applications. First, we introduce deformable substrates with structural stretchability, achieved by incorporating buckling and Kirigami structures into plastic films, and we systematically analyze the tensile deformation characteristics based on design elements. In addition, we examined intrinsically stretchable elastomeric substrates, which have gained considerable attention due to recent advances in material and processing technologies. Their spatial modulus patterning is studied by applying optimized design principles, achieved through network alignment and crosslinking control in homogeneous elastomers, as well as by incorporating heterogeneous structures within the elastomer materials. Finally, we discussed state-of-the-art stretchable display applications employing strain-engineered stretchable substrates, focusing on advantageous materials and structures based on the display components, processes, and target deformation characteristics. Building on this foundation, we discuss the development of next-generation free-form displays and aim to contribute to their application in various static and dynamic deformation environments.

Keywords: Strain-engineering methods, structurally designed plastic films, modulus-patterned elastomers, stretchable displays

INTRODUCTION

Displays play a crucial role in the industry, serving as a key component that enables efficient communication between humans and machines by delivering visual information[1,2]. Given that humans heavily rely on vision to process information, displays are essential for conveying this information accurately and efficiently[3,4]. With the rapid advancement of communication technology, displays have become indispensable in nearly every aspect of daily life, including smart homes, wearable devices, automotive systems, and digital signage[5-8]. As these diverse application areas continue to expand, research and development efforts are increasingly focused on creating displays with broader functionalities and innovative form factors. The development of flexible displays has been made possible by advancements in material and process technologies, leading to the commercialization of foldable and rollable displays with adjustable screen sizes, which have gained significant popularity among consumers due to their attractive designs[9-11]. Additionally, the accumulated expertise in display materials and processes, along with advancements in analyzing stress and strain during deformation, has further fueled research into displays with cutting-edge multi-form factors[12-14]. Particularly, stretchable displays, which can freely change shape as the device stretches, are regarded as the pinnacle of display form factor innovation and are expected to play a vital role in the future evolution of display technology[15,16].

In the effort to expand display form factors, extensive research and development have been conducted on substrates, electrodes, light-emitting elements, and driving components, with substrate materials consistently being a top priority[17-21]. For instance, flat panel displays have traditionally relied on glass substrates that offer high transmittance along with excellent thermal durability[22,23]. On the other hand, the emergence of flexible and foldable displays has shifted the focus toward researching thin and flexible glass and plastic substrates, which are better suited to handle the mechanical demands of bending and folding[24,25]. In this manner, different substrates are selectively utilized based on the display's target form factor. The selection process carefully considers not only the physical and chemical properties of the display materials but also the temperature and pressure requirements during processing, as well as the functional and aesthetic needs of the final application area. Therefore, to successfully realize display devices, it is crucial to balance these factors and choose the most suitable substrate.

This review provides a comprehensive overview of strain-engineered substrate materials and their applications in stretchable displays. It specifically categorizes and explains the differences between plastic films with structural stretchability and elastomers with intrinsic stretchability [Figure 1]. For plastic films, the review delves into the buckling and Kirigami structures that are commonly used to impart structural stretchability, providing detailed explanations of the deformation behaviors that arise from various design elements of these structures. For elastomers with intrinsic stretchability, it is emphasized that various deformation dynamics can be programmed under the same tensile input through spatial modulus patterning. This patterning can be achieved by aligning the elastomer network, adjusting the crosslink density, or creating structures on the substrate surface. Additionally, this can be accomplished by structurally designing heterogeneous materials, embedding them into the elastomer matrix, or forming patterns on the matrix surface with heterogeneous materials. Finally, the review explores the utilization of these strain-engineered stretchable substrates in display applications, adapted to different usage scenarios and environmental conditions, by examining various examples.

STRUCTURALLY DESIGNED PLASTIC SUBSTRATES

To provide stretchability to flexible plastic films, strategies are commonly used that incorporate structures capable of accommodating dimensional changes due to tensile deformation. To achieve this, various structural designs have been introduced, including serpentine structures with zigzag patterns[26,27], helical

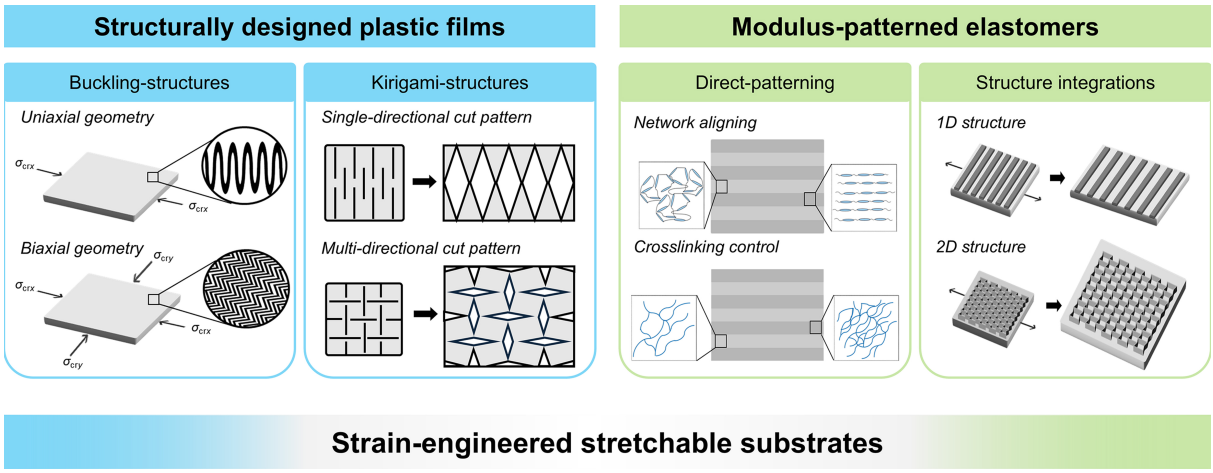

Figure 1. Overview of strain-engineered stretchable substrates for applications in display devices: structurally designed plastic films and modulus-patterned elastomers.

spring forms[28,29], accordion-like structures that enable repeated folding and unfolding[30,31], buckling structures that form wrinkles or wave-like patterns on substrate surfaces[32-35], and Kirigami designs, where cut patterns create voids upon deformation[36-39]. In this review paper, the focus is on buckling and Kirigami structures, which can be readily integrated into 2D planar substrates and provide sufficient effective area for device fabrication, especially in display applications. Buckling structures are deformation phenomena that occur when compressive forces are applied to plastic films, resulting in vertically oriented buckling structures on the surface once the critical compressive stress is exceeded[32-35]. By intentionally designing these structures, external deformation can be absorbed and dimensional changes accommodated, converting vertical deformation into horizontal expansion and thereby granting stretchability to flexible plastic films [Figure 2A]. Kirigami is a design technique that imparts stretchability to plastic films through the use of cut patterns[36-39]. Cut sections open under external forces, creating voids that allow for dimensional adjustments, while the size and orientation of the cut patterns are modified to precisely control stretchability and deformation behavior [Figure 2B]. As previously mentioned, this review paper explores the significance and potential of design strategies centered on buckling and Kirigami structures to enable strain programming in plastic films for applications in free-form displays.

Buckling-structured plastic films

The strategy for imparting stretchability to plastic films using buckling structures focuses on adjusting compressive stress beyond the critical threshold under various physical property conditions of the plastic film to program vertical buckling structures, thereby inducing the desired stretchable deformation. The critical compressive stress (σ_{cr}) refers to the specific stress condition at which the surface of the plastic substrate can no longer remain flat and begins to form vertical buckling structures. It is determined by the film's physical properties and dimensional parameters, such as Young's modulus (E) and thickness (t), and is defined as follows[32].

$$\sigma_{cr} = c\frac{E}{(1-\nu^2)}\left(\frac{t}{b_{cr}}\right)^2 \quad (1)$$

where c represents the buckling constant, b_{cr} denotes the half-width of the buckling structure formed under σ_{cr}, and ν refers to the Poisson's ratio of the plastic film. When a compressive stress (σ) exceeding σ_{cr} is applied to the film, the shape and size of the buckling structure are determined according to the applied σ conditions, thereby defining the stretchable properties of the film substrate. At this time, the amplitude (A)

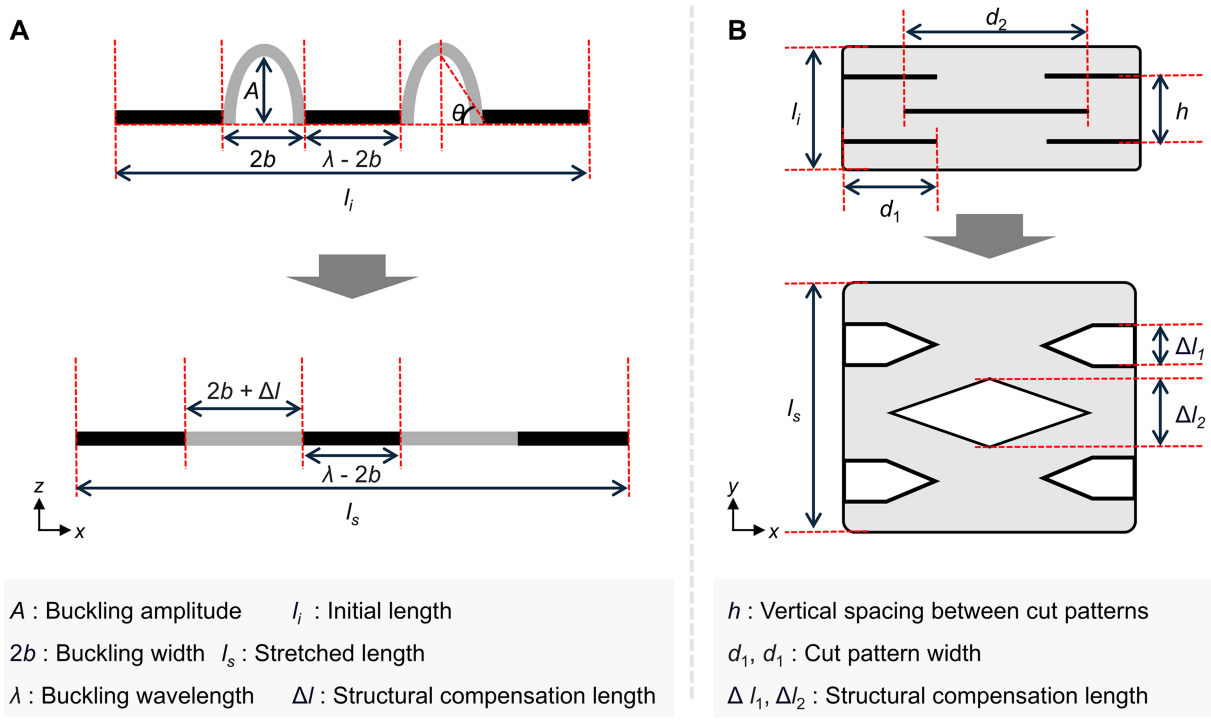

Figure 2. Dimensional compensation mechanism under tensile strain in (A) buckling and (B) Kirigami structures.

of the buckling structure can be summarized as follows[32].

$$A = t\sqrt{\frac{4}{3}\left(\frac{\sigma}{\sigma_{cr}} - 1\right)} \tag{2}$$

When buckling structures on the film surface have wider widths ($2b$), larger amplitudes (A), and shorter distances between them (λ - $2b$, where λ is the buckling period), the in-plane dimensional compensation during tensile deformation increases, thereby enhancing the substrate's stretchability. However, achieving such structures requires higher σ, which can cause more damage to the film during the buckling formation process and potentially reduce structural stability. Additionally, increasing the amplitude in buckling structures of the same width leads to a larger angle (θ), as shown in Figure 2A, which compromises mechanical reliability under repeated deformation and makes performance degradation or device delamination due to substrate deformation more likely in device applications. Therefore, by carefully designing the buckling structures considering the deformation behaviors and required stretchability of the target applications, an optimal balance between stretchability and structural stability should be achieved.

Various buckling structures can be achieved depending on the compression stress pattern applied to the plastic film, broadly categorized into uniaxial and biaxial buckling structures [Figure 3]. Methods for applying compression stress to a plastic substrate include attaching a pre-fabricated mold to the film and applying heat or pressure[40], directly scanning the substrate surface with a laser[41,42], or attaching the plastic film to a pre-stretched substrate and subsequently releasing it[43,44]. Uniaxial buckling structures arise when σ is applied along a single, specific axis of the plastic film, typically forming basic structures that facilitate stretchable deformation in the direction of compression [Figure 3A]. Positional variations in the magnitude of σ can occur due to material non-uniformity based on the film's boundary conditions or positional differences, generally resulting in the formation of aperiodic uniaxial buckling structures with variable

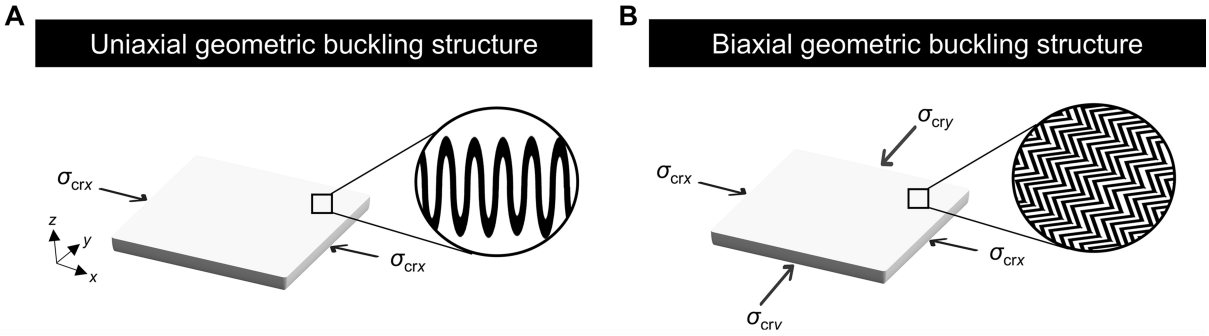

Figure 3. Schematic diagrams of (A) uniaxial geometric and (B) biaxial geometric buckling structures under unidirectional compressive stress and multi-directional compressive stress, respectively.

wavelengths and amplitudes across different locations on the film[45-49]. Drack *et al.* developed an ultrathin stretchable polyethylene terephthalate (PET) film with anisotropic wrinkle structures through a pre-strain process[45]. They achieved a film with up to 140% stretchability by attaching a pre-strained elastic adhesive tape to a PET film embedded with electronic devices and then detaching it to induce aperiodic buckling, providing mechanical stability that allowed the device to continue functioning under various deformations such as crumpling and twisting [Figure 4A]. Similarly, Li *et al.* formed an aperiodic buckling structure with an average peak-to-peak distance of 100 μm and a curvature radius of 70 μm by attaching a polyimide (PI) film coated with Ag nanowires (AgNWs) to a pre-strained adhesive film and then releasing it[47]. This method demonstrated excellent electrical performance even under tensile conditions, maintaining consistent luminance despite repeated stretching when applied to display devices [Figure 4B]. Additionally, Mishra *et al.* created an aperiodic buckling structure by attaching a polystyrene (PS) film to a polydimethylsiloxane (PDMS) substrate, bending the substrate, and then releasing it to tailor the stress[49]. This resulted in the highest buckling density at the center of the substrate, decreasing towards the edges, and their study demonstrated that the buckling structures not only imparted stretchability but also induced changes in the light diffusion performance through the substrate [Figure 4C].

Aperiodic buckling structures can cause non-uniform local deformations at different positions of the substrate during stretching, leading to stress accumulation in specific areas during repeated deformation and mechanical stability issues in device applications, which can hinder their practical use in electronic devices[50]. To address these challenges, methods have been proposed to induce uniform compressive stress in the film using pre-designed molds or laser scanning[51-53]. Zhao *et al.* replicated periodic buckling structures on PI films using an isotropic mold with a period of 200 μm and an amplitude of 160 μm[51], applying the resulting PI film as a temperature sensor capable of detecting dynamic temperature changes on the skin [Figure 4D]. Yin *et al.* employed laser patterning to form periodic buckling structures on an elastic stamp in a non-contact manner, then transferred these structures onto a stretched PET film to create uniform buckling structures[52]. This approach led to the development of a plastic film with up to 100% stretchability and spatially uniform deformation, which was subsequently applied to organic light-emitting diode (OLED) devices [Figure 4E]. Hartmann *et al.* utilized chemical vapor deposition (CVD) to precisely deposit a parylene-C film onto a silicon mold with pre-fabricated micro-structures, having a peak spacing of 50 μm. Afterward, they added Cr/Au layers to the film to impart electrical conductivity, followed by an additional layer of parylene for complete encapsulation[53]. The resulting buckled plastic film demonstrated structural stretchability, withstanding tensile strains of up to 55%. Due to the excellent insulating properties and biocompatibility of parylene-C, this buckling-structured conductive film is well-suited for both wearable devices and stretchable electronics, such as bioelectronic devices, providing enhanced durability

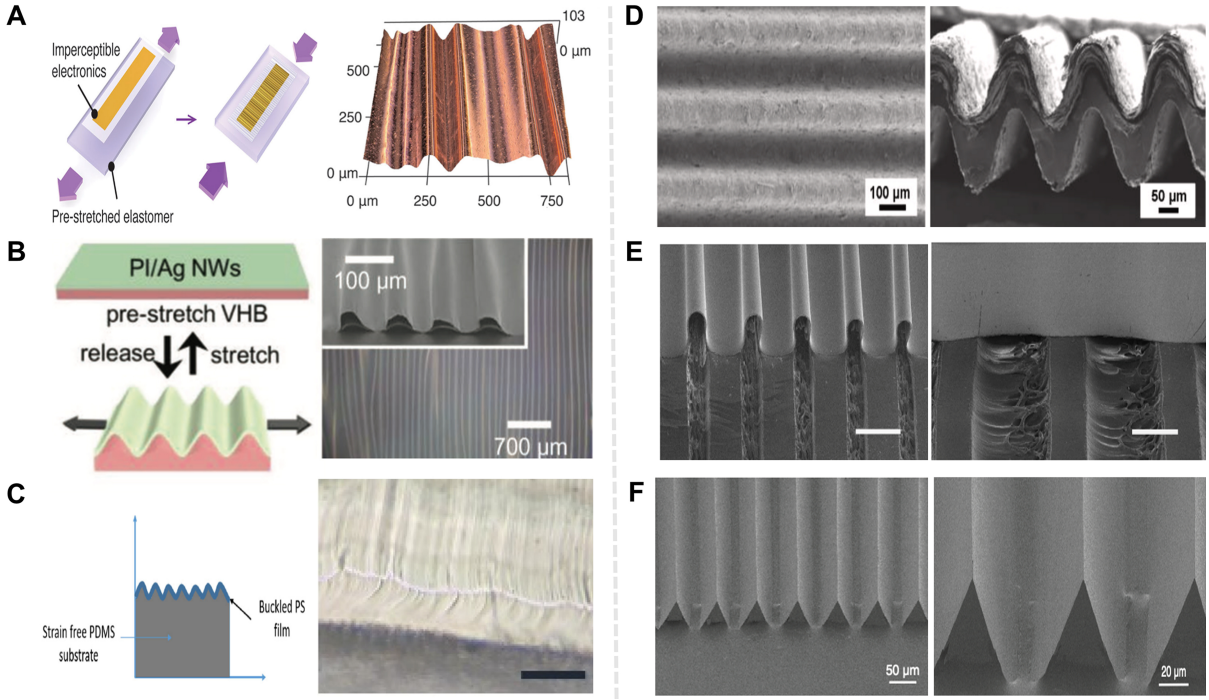

Figure 4. Flexible films with vertically uniaxial buckling structures. (A) Schematic of a PET film with an aperiodic buckling structure and attached electronic foil, along with a 3D digital optical microscope image. Reproduced with permission[45]. Copyright 2015, Wiley-VCH; (B) Schematic of the buckling structure formed on a PI/AgNW composite electrode and the wrinkled surface morphology of the composite electrode. Reproduced with permission[47]. Copyright 2019, Wiley-VCH; (C) Schematic of the buckling structured PS film on the PDMS, including a cross-sectional depiction of the interface layer between the PS and the PDMS. Reproduced with permission[49]. Copyright 2024, Springer Nature; (D) Planar SEM image of a PI film replicated using a micro-wrinkle pattern mold, including surface morphology and cross-sectional SEM images of the wrinkles. Reproduced with permission[51]. Copyright 2024, Elsevier; (E) SEM image of a periodic buckling pattern on a PET film created by a laser-programmable process. Reproduced with permission[52]. Copyright 2016, Springer Nature; (F) SEM images of the isotropically buckled parylene film structure replicated using a micro-patterned silicon mold. Reproduced with permission[53]. Copyright 2021, Wiley-VCH. PET: Polyethylene terephthalate; PI: polyimide; AgNW: Ag nanowire; PS: polystyrene; PDMS: polydimethylsiloxane; SEM: scanning electron microscope.

and performance [Figure 4F].

To enable diverse deformation behaviors in buckled plastic substrates, a method has been suggested that applies multiple σ of varying magnitudes and directions to the film, creating biaxial or multiaxial buckling structures [Figure 3B]. These structures can maintain similar dimensional and periodic characteristics as conventional uniaxial buckling structures while providing enhanced stretchability or multi-directional deformation potential[54-56]. Hyun *et al.* applied bidirectional mechanical strain to a PS film and then sequentially released the strain to create herringbone structures[54]. The formed herringbone structures evenly distribute stress across each direction, enhancing mechanical stability and allowing for greater compensation for deformation compared to uniaxial buckling structures [Figure 5A]. Additionally, different herringbone structures could be achieved depending on the magnitude of the bidirectional σ, with the grooves of the herringbone structure filled with a polymer and silver nanoparticle mixture to create a stretchable conductive electrode. Yu *et al.* developed a stretchable electrode by fabricating a super-aligned carbon nanotube (SACNT) film with a wrinkled structure using a biaxial pre-strain method, allowing it to withstand large strains from multiple directions repeatedly[55]. They also created a stretchable SACNT/activated-carbon supercapacitor using activated-carbon powder and SACNT. The circuit, which integrates the stretchable electrode with the SACNT/activated-carbon supercapacitor, demonstrated excellent reliability and high-capacity performance under significant strain in various directions, indicating

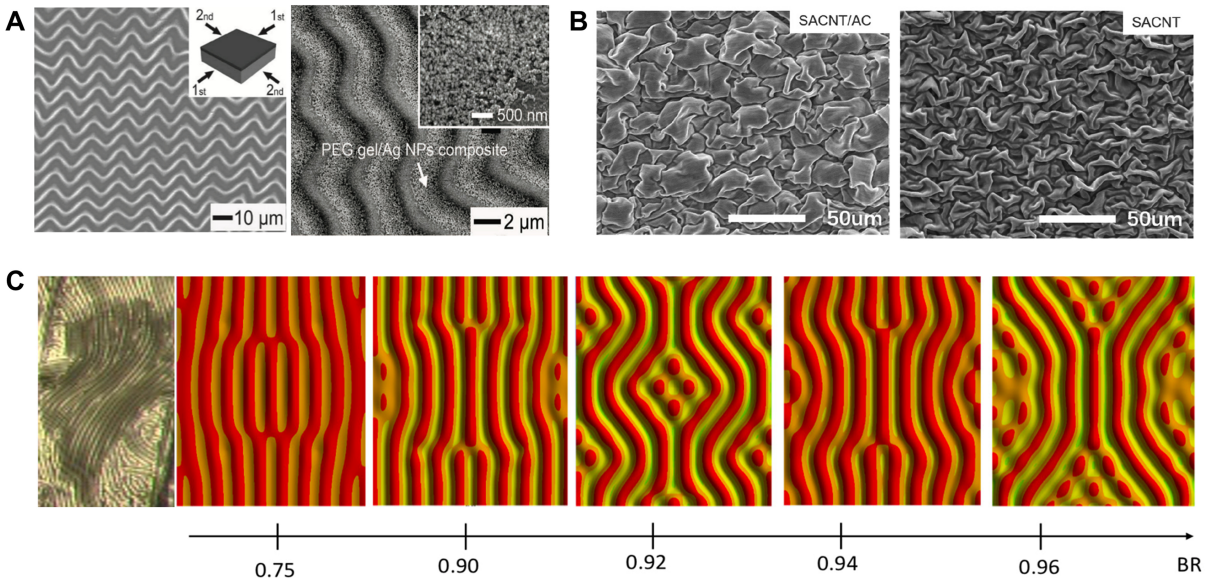

Figure 5. Flexible films with vertical multiaxial buckling structures. (A) 2D herringbone pattern created through sequential tensile release and an SEM image of a polymer gel/metal nanoparticle composite. Reproduced with permission[54]. Copyright 2011, Wiley-VCH; (B) Randomly buckled SACNT/AC supercapacitor and SACNT conductor films formed using a biaxial pre-strain technique. Reproduced with permission[55]. Copyright 2020, Royal Society of Chemistry; (C) Simulation results showing changes in buckling structures according to the *x*-*y* BR (the left image displaying the buckling structure at BR 0.75). Reproduced with permission[56]. Copyright 2024, MDPI. SEM: Scanning electron microscope; SACNT: super-aligned carbon nanotube; AC: activated-carbon; BR: biaxial ratio.

promising potential for various stretchable electronic device applications [Figure 5B]. Seok *et al.* applied isotropic or anisotropic σ along two mutually perpendicular axes to introduce buckling structures onto a PI substrate with transferred Au[56]. They analyzed the regularity of the buckling structures based on the differences in σ applied to the two axes and adjusted these parameters to modify the density and shape of the buckling structures, enabling their application in stretchable optical devices that regulate light scattering and transmission [Figure 5C].

Kirigami-structured plastic films

Plastic films with Kirigami structures expand at the cut sections under tension, creating empty spaces that accommodate dimensional changes and provide stretchability. This deformation behavior is determined by the size, orientation, and combination of the cut structures, while physical properties such as the film's modulus (E) and thickness (t) also play a crucial role in influencing its tensile performance[57,58]. Therefore, understanding the interaction between these factors is essential for designing Kirigami-structured plastic films with customized deformation properties tailored to specific applications. The formation of Kirigami cut patterns on plastic films is primarily achieved through etching processes using pre-designed masks or by laser machining[59-63]. In particular, recent advancements in high-frequency laser technology have made it possible to create intricate and precise patterns on plastic films while minimizing thermal damage, significantly improving manufacturing efficiency[61,62]. Among various cut patterning techniques, scanning laser cutting stands out, as it enables the laser beam to move rapidly across the entire film, allowing for the swift and efficient creation of the desired patterns[63,64]. This technology is especially advantageous in mass production environments where large films must be processed in a short time, making it efficient in terms of both time and cost. Furthermore, the introduction of various Kirigami structures not only facilitates simple unidirectional tensile deformation but also allows for 2D or 3D spatial expansion depending on the arrangement and combination of cut patterns.

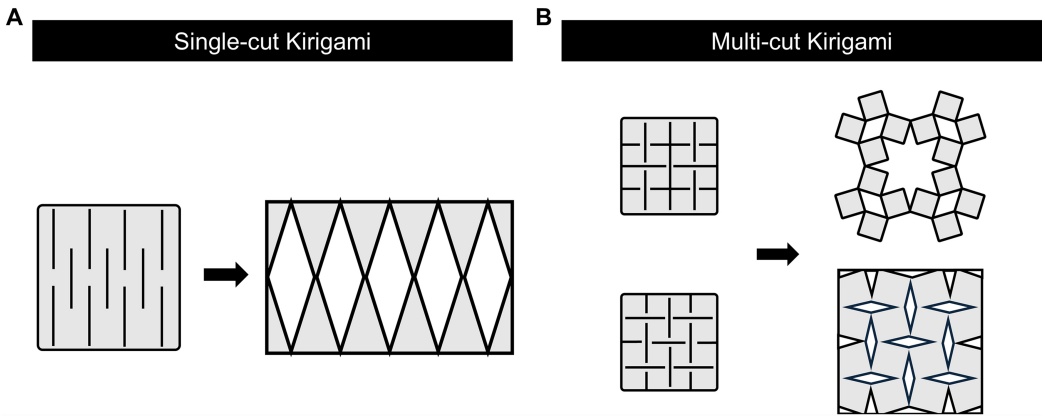

Figure 6. Schematic diagrams of plastic films with (A) single-cut and (B) multi-cut Kirigami structures, showing before and after stretching.

In single-directional expansion Kirigami structures, the tensile direction and the pattern direction are aligned perpendicularly to each other [Figure 6A]. While reducing the spacing between the cut patterns or increasing their length and width can significantly enhance the stretchability of plastic films, it also tends to increase structural distortion during deformation. Therefore, to balance stretchability and structural stability effectively, it is essential to meticulously design the pattern and precisely tailor the film's strain characteristics to the desired tensile range and configuration, thereby achieving optimal performance. Lee *et al.* utilized high-frequency laser technology to create precise cutting patterns on PET films while minimizing thermal damage[65]. As a result, the plastic film cut with a 10:1 length-to-gap ratio maintained excellent performance under tensile strains exceeding 200%, as well as under twisting and bending deformations [Figure 7A]. Hwang *et al.* developed a hybrid Kirigami-bridge structure to further enhance the single-directional stretchability of plastic films[66]. This pattern combines long, precisely designed major cuts in the transverse direction with short minor cuts in the longitudinal direction [Figure 7B]. The addition of minor cuts significantly increased the stretchability from 200% to 330%, and the free movement of the Kirigami domains also improved flexibility, making the films more suitable for curved surface applications. Similarly, Guan *et al.* applied a Kirigami structure with an increased aspect ratio of 20:1 to PET films, targeting applications that require high stretchability[67] [Figure 7C]. By transferring thin nanosheets onto the film, they enhanced its resistance to tensile deformation. This approach demonstrates both high stretchability and improved mechanical strength, highlighting the potential for use in advanced applications that require both flexibility and durability. Won *et al.* developed a transparent Kirigami electrode (TKE) film by applying high-power nanosecond laser ablation technology to a clear polyimide (cPI) film layered with AgNW, thereby achieving both excellent stretchability and high optical transparency[68]. The resulting TKE film was utilized as a wearable heater, maintaining stable performance even under strains of up to 200% while providing uniform heat distribution [Figure 7D]. These superior properties make it highly suitable for a variety of wearable devices that require localized thermal management, such as electronic skin (E-skin) for wound healing and body temperature maintenance.

The multi-directional expansion Kirigami structure involves combining cutting patterns with various axes and dimensions on a plastic film, allowing the designed voids to expand simultaneously in multiple directions during substrate stretching [Figure 6B]. This design hierarchically structures factors such as the angles between patterns, dimensional variations, and the positions of intersections, enabling the pattern domains to achieve multi-directional expansion through linear or rotational movements during deformation[69,70]. This design approach enables various types of deformation and ensures stability even in

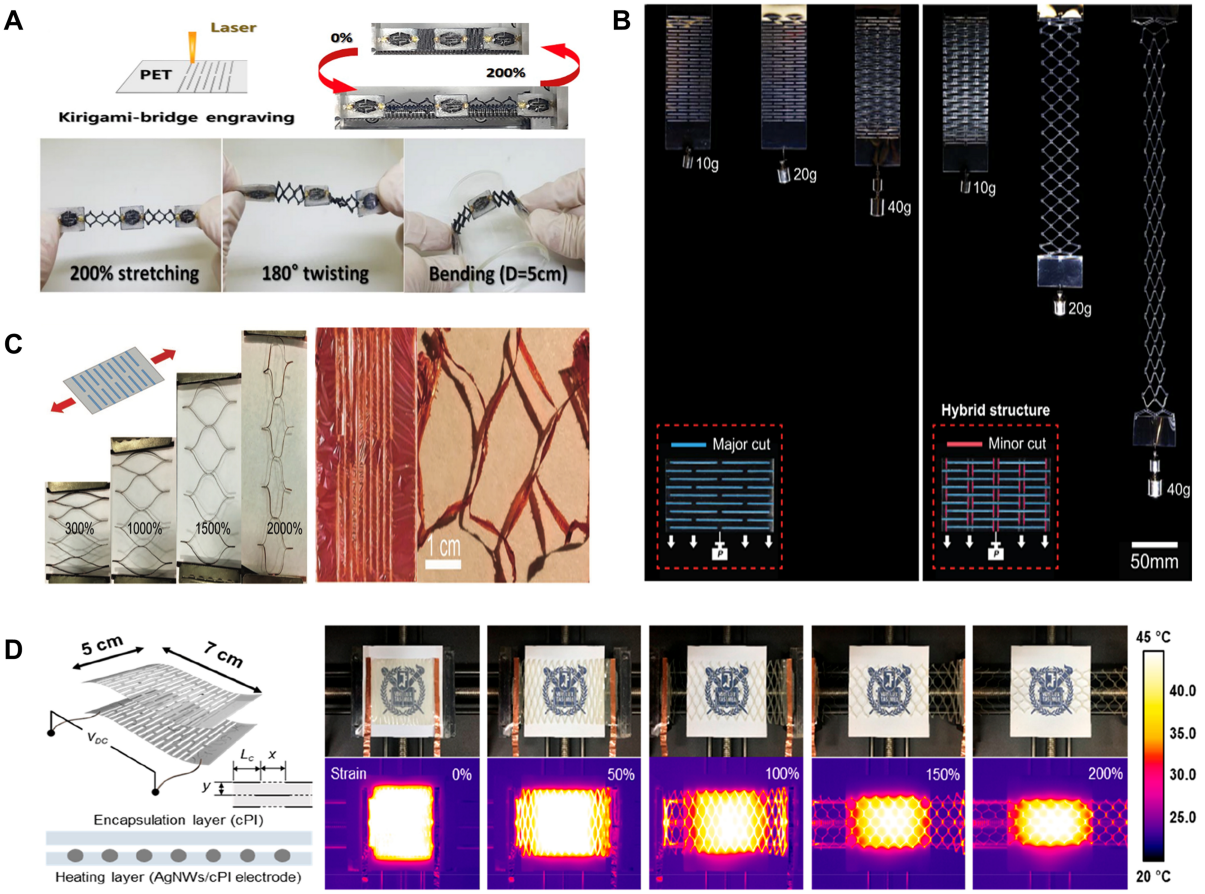

Figure 7. Flexible films with single-directional stretchability incorporating Kirigami patterns. (A) PET film with a single-cut Kirigami pattern, demonstrating tensile deformation up to 200%, along with twisting and bending. Reproduced with permission[65]. Copyright 2020, Elsevier; (B) Photographs of a Kirigami pattern with only major cuts and a hybrid pattern featuring both major and additional cuts. Reproduced with permission[66]. Copyright 2018, Springer Nature; (C) Images of the Kirigami pattern deformed under various strains and a freestanding nanosheet. Reproduced with permission[67]. Copyright 2018, Wiley-VCH; (D) Electrothermal properties tested under tensile conditions from 0% to 200% strain, using a heating layer formed on a uniaxially cut cPI film substrate. Reproduced with permission[68]. Copyright 2019, American Chemical Society. PET: Polyethylene terephthalate; cPI: clear polyimide.

complex movements, thereby expanding the potential applications of stretchable substrates in multi-modal robots and curved optical devices[71,72]. An *et al.* demonstrated that introducing hierarchical cuts into flat Kirigami sheets can enhance the mechanical response of the substrate[73]. By adjusting the parameters of the cuts and the thickness of the film, they effectively programmed the stress-strain response and expanded the range of mechanical responses achievable by combining various hierarchical structures, allowing for the encryption of information within the structure [Figure 8A]. Khosravi *et al.* proposed a pneumatic actuator with pre-programmed motion capabilities using a composite Kirigami-structured substrate[74]. The Kirigami skin, wrapped around a cylindrical balloon, transformed volumetric expansion from pneumatic pressure into anisotropic stretching and shearing, resulting in a combination of axial extension and rotational deformation of the actuator. The two groups of slit cuts in the Kirigami skin imparted nonlinear kinematic and mechanical properties, allowing the combined control of extension and rotational movements by adjusting the angles and lengths of the slit cuts [Figure 8B]. Rafsanjani *et al.* proposed a methodology for designing deformation behaviors based on the correlation between groups of Kirigami cut patterns, analyzing the mechanical deformation behavior resulting from different combinations of cuts on a square thin plastic sheet through experimental, analytical, and numerical studies[75] [Figure 8C]. This methodology

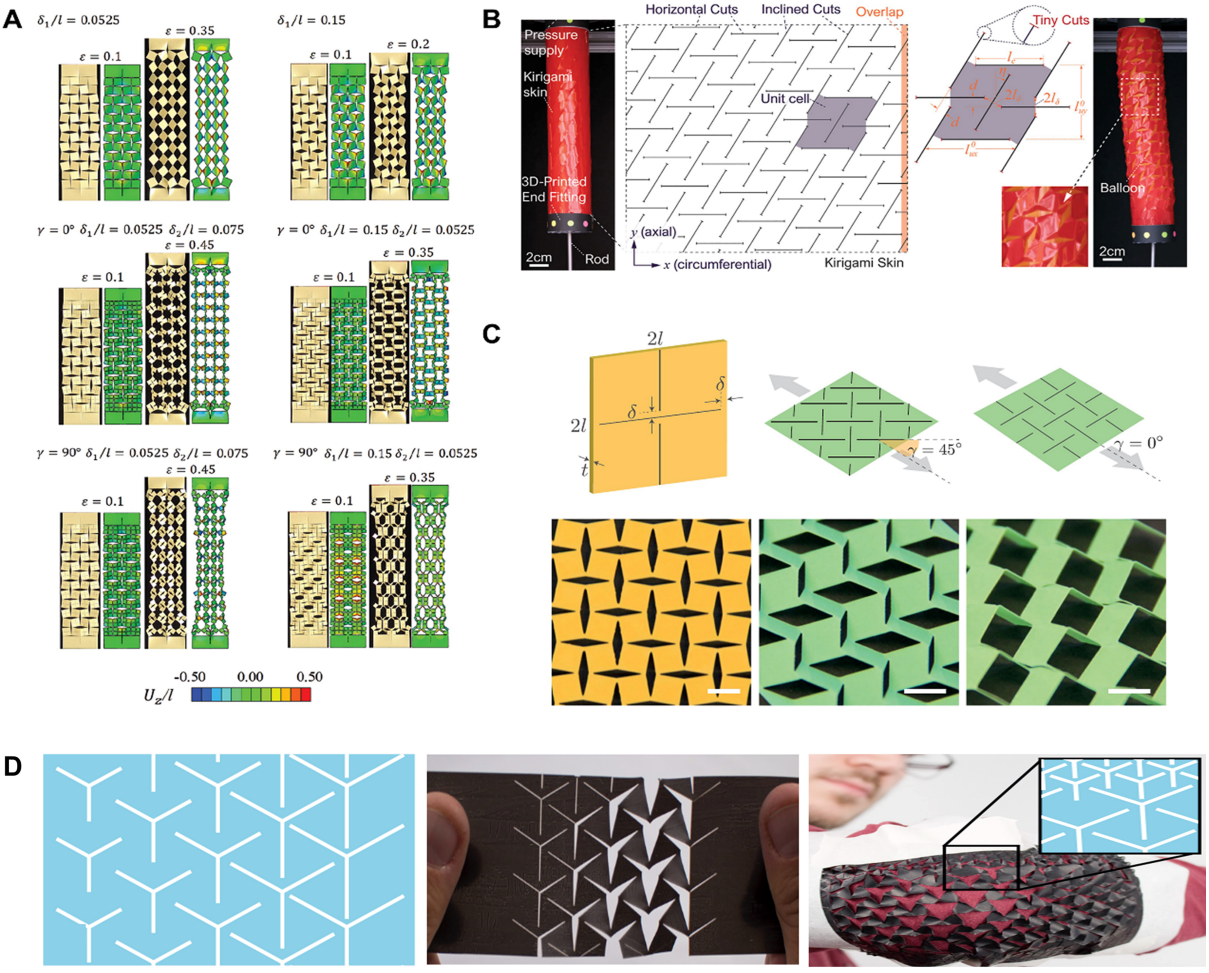

Figure 8. Flexible films with multi-directional stretchable Kirigami patterns. (A) Kirigami sheet with stress-strain characteristics programmed based on the parameters of the cut patterns and the thickness of the film. Reproduced with permission[73]. Copyright 2019, Wiley-VCH; (B) Pneumatic actuator with pre-programmed motion capabilities using a composite Kirigami-patterned substrate. Reproduced with permission[74]. Copyright 2021, Khosravi, Iannucci and Li; (C) Mechanical deformation of Kirigami sheets resulting from various combinations of cut patterns on a square flexible sheet. Reproduced with permission[75]. Copyright 2017, American Physical Society; (D) Tensile deformation of Y-cut PET film and application of a textile sensor patch, developed based on the designed film, to a bent elbow. Reproduced with permission[76]. Copyright 2019, Association for Computing Machinery. PET: Polyethylene terephthalate.

not only offers a straightforward approach to producing Kirigami sheets but can also be combined with optimization techniques to design perforation patterns that generate complex shapes under external loads. This approach can be applied to create Kirigami sheets across various scales, from transformable meter-scale structures to tunable nanoscale surfaces, providing a simple way to manufacture complex, morphable structures from flat perforated sheets. Additionally, Groeger *et al.* ingeniously combined Y-shaped cut patterns to develop a PET film with significantly enhanced adhesion to complex curved surfaces[76] [Figure 8D]. The Y-shaped cut pattern, created through laser patterning, provides anisotropic stretchability, allowing the film to flexibly adapt to intricate skin contours and dynamic movements. This pattern evenly distributes the stress generated when the film stretches, ensuring structural stability. As a result, the film adheres securely to the skin, even during user movement, making it ideal for applications such as E-skin and wearable devices.

MODULUS-PATTERNED ELASTOMERIC SUBSTRATES

In the previous section, we explored the introduction of various structures into plastic substrates to impart stretchability and examined the effects of different structural design approaches on deformation. These structured plastic films possess mechanical and optical properties suitable for display device applications, yet a challenge remains in balancing the substrate's stretchability with the effective area needed for device formation[77]. Additionally, achieving stretchability often requires adding compensatory structures perpendicular to the surface and expanding void spaces, which may compromise reliability in applications requiring complex shape deformation, thereby limiting their practical application range. Recent advancements in low-temperature and printing processes have enabled the formation of devices at relatively low temperatures, leading to increased exploration of using inherently stretchable elastomer materials as substrates[78-81]. Elastomers can undergo tensile deformation without the need for compensatory structures, as the chains within the matrix extend, and the elastomer matrix, which forms a random network, allows for tensile deformation in all directions[79]. Researchers have recently been exploring strain engineering techniques that spatially pattern the modulus of elastomers to address unwanted deformations in traditional elastomer substrates (e.g., unnecessary contraction due to the Poisson's effect) or to achieve multi-directional dynamic tensile deformation instead of unidirectional static stretching[82,83]. This section examines methods for controlling the deformation behavior of elastomer substrates by spatially patterning the modulus, including the regulation of molecular alignment and crosslink density in homogeneous elastomers or the incorporation of heterogeneous structures.

Direct-patterning on homogeneous elastomer substrates

To control the dynamic behavior of homogeneous elastomer films, one effective method is to pattern the modulus of the film by spatially reconfiguring the polymer chain network. This approach involves aligning the molecules that constitute the elastomer network in specific directions or selectively controlling the crosslink density of the network at different locations[84,85] [Figure 9]. Key methods for controlling molecular alignment in elastomer networks include introducing spatially oriented rubbing patterns on the mother substrate surface where the film is fabricated, guiding molecules - especially those with anisotropic structures - to align in the desired direction[86]. Additionally, techniques such as applying mechanical stretching to the substrate to rearrange molecules through shear forces[87,88] and using directional electromagnetic fields to control molecular orientation can also be utilized[89]. By physically aligning polymer chains in a specific direction, differences in modulus arise between the aligned and non-aligned directions, and designing and patterning this alignment allows for various deformations to be induced even under the same external tensile input [Figure 9A]. Among various elastomer materials, these molecular alignment techniques are commonly used in block copolymer (BCP) elastomers[90-93] with chemical incompatibility between molecules or liquid crystal elastomers (LCEs)[94-100] that have anisotropic rigid domains.

BCPs are polymer chains made up of two or more monomers with unique properties, and due to the chemical incompatibility between these monomers, phase separation occurs in response to external stimuli, resulting in alignment[90]. This alignment generally leads to modulus anisotropy along specific axes and is primarily achieved through mechanical or chemical methods. Ye *et al.* investigated the effects of unidirectional alignment of polystyrene-block-polydimethylsiloxane (PS-b-PDMS) thin films on their macroscopic deformation and mechanical properties[91]. By utilizing the cold zone annealing-soft shear (CZA-SS) technique, they successfully aligned PDMS domains within the PS matrix [Figure 10A]. This alignment resulted in a significant increase of up to 31% in the elastic modulus along the alignment direction, causing notable deformation anisotropy in the film's mechanical behavior. Park *et al.* employed a solvent annealing method using a chamber with solvent and a heater to induce the self-assembly of BCPs, leading to the successful reorganization and alignment of domains within the film[92] [Figure 10B]. This

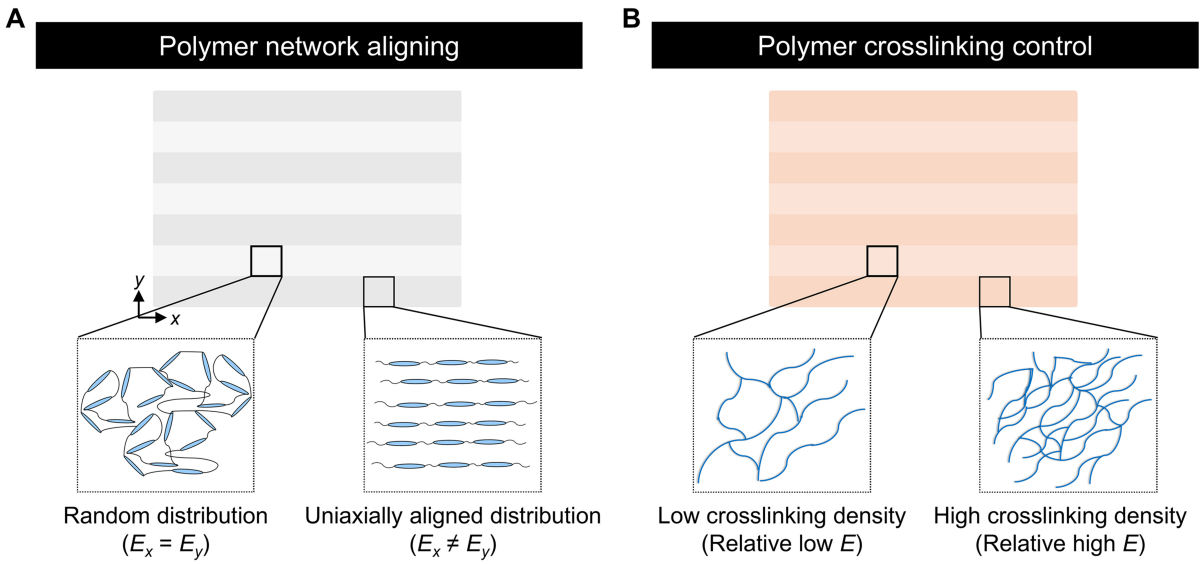

Figure 9. Schematic diagrams of homogeneous elastomer films directly patterned by (A) aligning the polymer network and (B) adjusting the crosslink density.

method operates at low temperatures below 100 °C and can produce highly ordered sub-10 nm patterns in under a minute, generating uniform and low-defect patterns that are effective for various BCP morphologies. Ditte *et al.* developed a triblock copolymer (TBC) combining poly-diketopyrrolopyrrole-thienothiophene (PDPP-TT) with PDMS to achieve the low elastic modulus and high charge mobility essential for stretchable electronics[93] [Figure 10C]. Through the arrangement of this TBC, they induced anisotropic charge transport within the elastomer film, leading to performance variations based on the direction of the electric field and the applied strain. Grazing-incidence wide-angle X-ray scattering (GIWAXS) analysis revealed that the TBC has an edge-on molecular orientation, aligned perpendicular to the substrate, which facilitates charge transport pathways parallel to the film surface and enhances electron mobility.

LCEs consist of rigid liquid crystal (LC) mesogens crosslinked within a polymer network, and the orientation of this network is determined by the direction of the LC director[94]. The van der Waals interactions between the LC directors allow for hierarchical alignment in the thickness direction. Depending on the anchoring properties, alignment directions, and positional alignment profiles of mechanically or optically aligned thin films, various geometries such as vertical, horizontal, and twisted configurations can be designed, making it highly advantageous for inducing multi-directional deformation[95]. Mechanical alignment involves physically rubbing the surface of the alignment layer to guide the alignment of LC directors, providing strong anchoring properties that are effective even for thick films[96,97]. However, it has limitations in inducing complex spatial alignments. In contrast, optical alignment involves exposing the alignment layer to specific wavelengths of light, which induces chain cleavage and recombination to provide directional control[98,99]. By patterning the light source, spatial alignment can be easily controlled, making it more suitable for complex alignments. Choi *et al.* developed a LC polymer designed to form a twisted geometry in the thickness direction and combined it with an azobenzene monomer to create an ultraviolet (UV)-responsive LCE strip[96] [Figure 10D]. By exposing the aligned film to light, they induced curvature deformation in the designed helices and applied this to a soft robot capable of programmable multi-motion. Ahn *et al.* applied inhomogeneous stretching and optical alignment techniques during the film fabrication process to induce patterned molecular orientation, creating LCE films with programmable complex

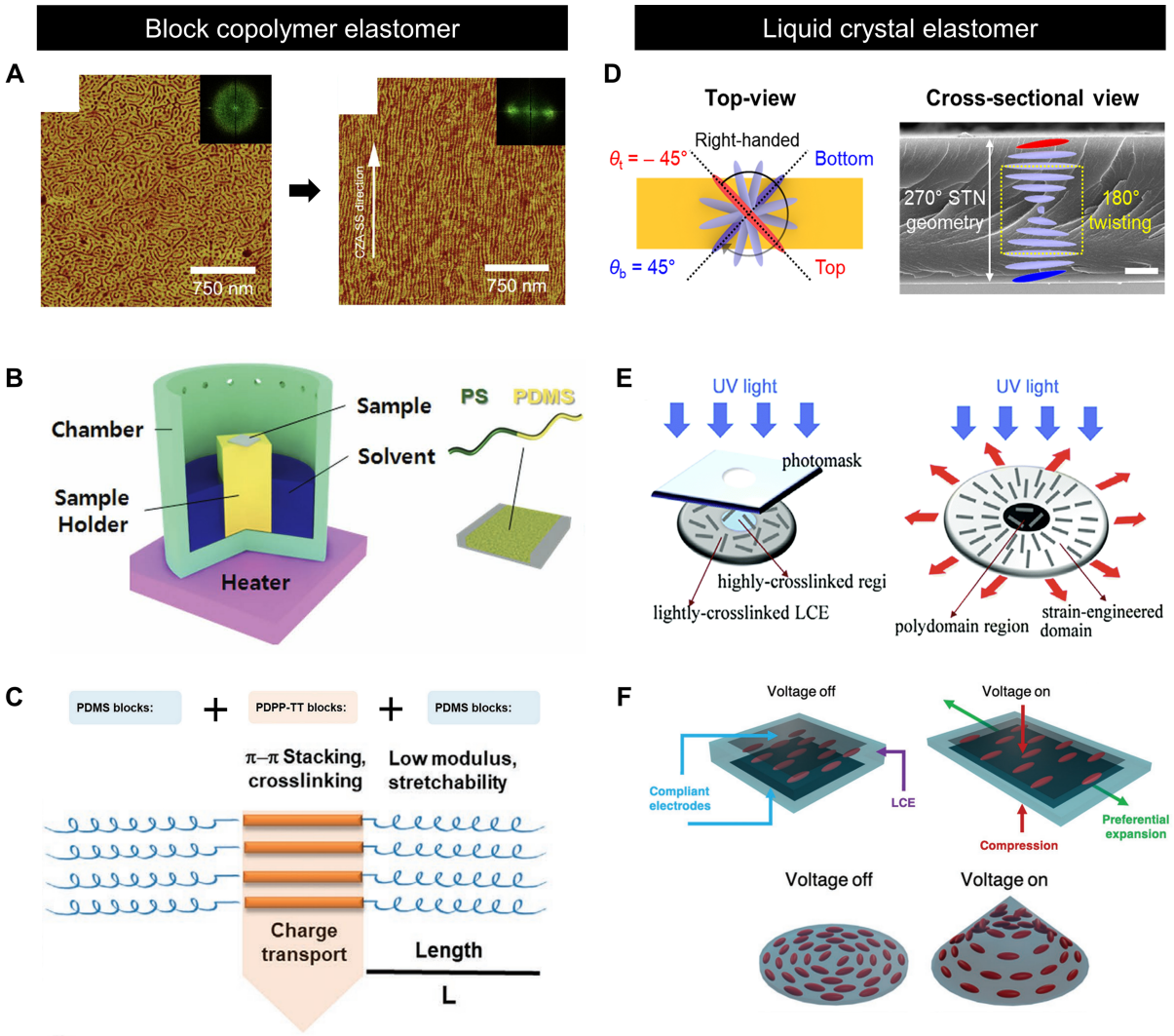

Figure 10. Homogeneous elastomers with spatially aligned polymer networks. (A) Domain images of the substrate surface showing the effects of unidirectional alignment in PS-b-PDMS thin films through the shearing process. Reproduced with permission[91]. Copyright 2013, American Chemical Society; (B) Self-assembled BCP elastomer film achieved through a solvent annealing process. Reproduced with permission[92]. Copyright 2012, Wiley-VCH; (C) Schematic of TBC composed of PDPP-TT and PDMS blocks, exhibiting anisotropic charge transport characteristics. Reproduced with permission[93]. Copyright 2021, Wiley-VCH; (D) LC polymer strip with a super-twisted nematic structure in the thickness direction. Reproduced with permission[96]. Copyright 2023, Wiley-VCH; (E) Programmable complex deformation LCE films created using non-uniform stretching and optical alignment techniques. Reproduced with permission[98]. Copyright 2015, Elsevier; (F) Schematic of anisotropic expansion of an LCE film with LC molecules aligned in a designed orientation under applied voltage. Reproduced with permission[100]. Copyright 2021, Wiley-VCH. PS-b-PDMS: Polystyrene-block-polydimethylsiloxane; BCP: block copolymer; TBC: triblock copolymer; PDPP-TT: poly-diketopyrrolopyrrole-thienothiophene; PDMS: polydimethylsiloxane; LC: liquid crystal; LCE: liquid crystal elastomer.

deformations, and applied these films to various electronic devices[98] [Figure 10E]. Fowler *et al.* demonstrated that LCEs exhibit excellent directional actuation in response to electric fields[100]. The directional modulus of LCEs can vary by up to 14 times depending on the alignment of LC molecules, and the dielectric constant changes according to the director orientation, enabling anisotropic expansion when an external voltage is applied [Figure 10F]. These materials achieve up to 20% strain as voltage is applied, demonstrating rapid deformation rates with minimal hysteresis. Furthermore, by applying spatial patterning to the LC director, they confirmed that precise transformation from 2D to 3D deformation is achievable.

Another approach to controlling deformation in homogeneous elastomers is by spatially patterning the crosslink density. Differences in crosslink density within the elastomer lead to changes in modulus, and by patterning these variations, the deformation behavior of the film can be controlled even under the same external tensile conditions [Figure 9B]. In elastomers, crosslinking occurs by supplying energy, such as heat or light, to form crosslink sites and create a 3D polymer network, depending on the types of monomers and crosslinkers. The crosslink density can be adjusted not only by the amount of external energy supplied but also by the content of crosslinkers within the material[101-106]. Methods for spatially patterning crosslink density include using patterned masks to selectively distribute thermal or light energy across the elastomer film or directly irradiating with patterned energy sources[101-104]. Notably, photo-crosslinking enables precise and complex pattern formation, allowing modulus patterning not only in-plane but also in the out-of-plane direction by adjusting penetration conditions according to wavelength within the elastomer matrix[105]. This crosslink density patterning approach makes it possible not only to regulate the overall mechanical behavior of a homogeneous elastomer substrate but also to enhance deformation resistance or flexibility in specific regions. Nauman *et al.* produced a film from a thermally cross linkable elastomer material containing photo-responsive color-changing dyes, patterning the crosslink density by generating spatial temperature variations using a gradient grayscale patterned film and an infrared light source[102]. When an external actuation light source was applied to the fabricated film, variations in photochromic dynamics appeared according to the crosslink density in different regions, and this technology was subsequently applied to artificial iris devices for contact lenses [Figure 11A]. Park *et al.* achieved a remarkable enhancement in the modulus of specific regions within a photo-cross linkable elastomer by up to 38,000% by selectively irradiating the material with patterned light[103] [Figure 11B]. This innovative selective patterning technique allowed them to precisely create distinct soft and hard regions within the film, facilitating the development of high-performance pressure sensors. These sensors are capable of maintaining exceptional sensitivity while effectively reducing the adverse effects of deformation, thus offering robust performance across various applications. Kang *et al.* developed a bio-based thermoplastic vulcanizate (TPV) by blending polyester elastomer (BPE) with polylactide (PLA), followed by a dynamic crosslinking process involving high temperatures and strong shear forces[104] [Figure 11C]. They adjusted the ratio of BPE to PLA to control the overall modulus of the TPV elastomer film. By optimizing the blend ratio and crosslinking process, they achieved optimal tensile strength and elongation at break for the TPV under external tensile conditions. This composite material exhibits high reprocessability and maintains its mechanical performance under repeated deformation, providing stable deformation behavior over an extended period.

Structural assembly of heterogeneous materials

The structural assembly of heterogeneous materials involves integrating pre-designed structures into an elastomer matrix to achieve desired deformations [Figure 12]. Unlike merely patterning homogeneous materials, this approach leverages the deformation control effects of assembled structures to more effectively manage both macroscopic and localized deformations, enabling in-plane and 3D deformation modes that broaden potential applications[107-116]. Heterogeneous material structures for controlling substrate deformation are manufactured through processes such as mold forming[109,111] and laser cutting[107,108], with recent widespread adoption of 3D printing-based additive manufacturing processes[116] further enhancing design flexibility by enabling the realization of complex structures.

The incorporation of 1D structures involves embedding linear elements such as fibers, rods, and ribbons into elastomers, where these components are predominantly aligned in a single direction to enhance axial deformation characteristics and control mechanical properties along that axis[109-112] [Figure 12A]. For instance, integrating frame-like structures or fibers into an elastomer substrate allows these elements to serve as mechanical supports that effectively control deformation when the substrate is stretched in a specific direction. Nauman *et al.* employed elastomer materials with high modulus to create a 1D line

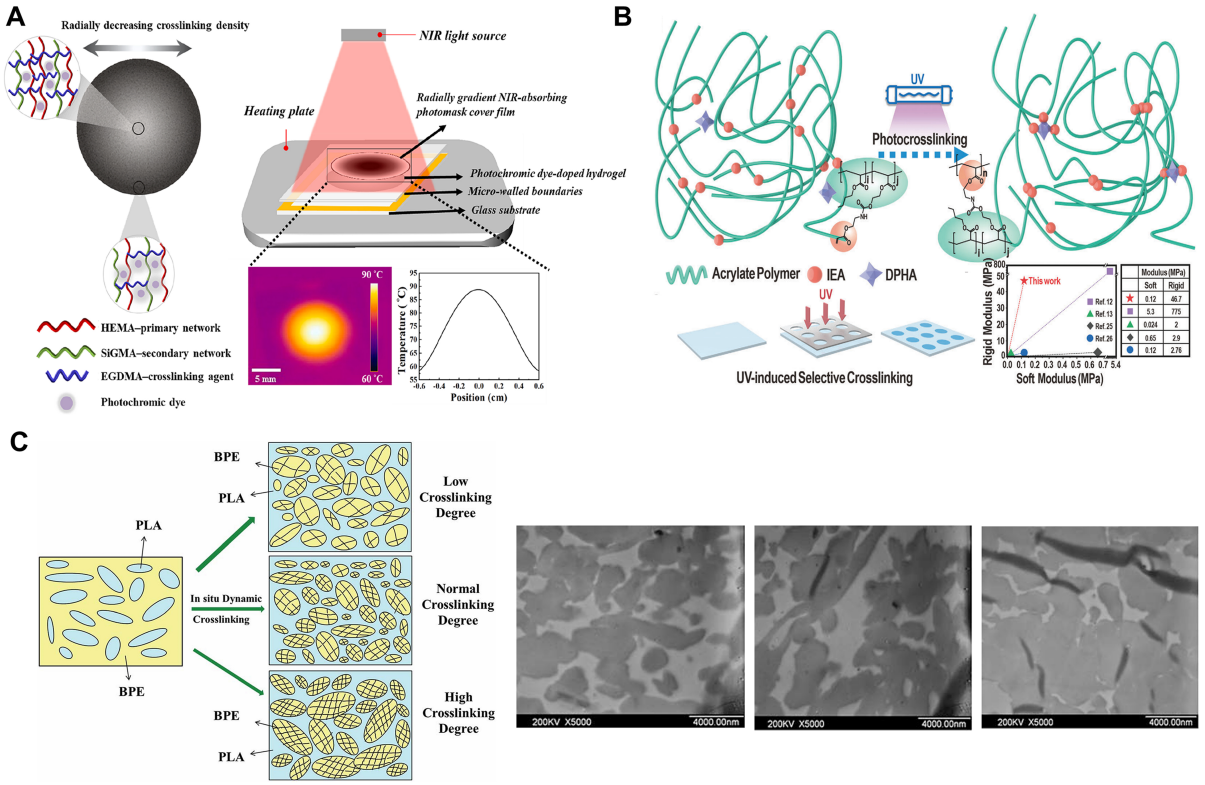

Figure 11. Homogeneous elastomers with patterned crosslink density. (A) Elastomer films with spatially patterned crosslink density achieved through a gradient thermal profile during the crosslinking process, along with an example of artificial iris application. Reproduced with permission[102]. Copyright 2024, Elsevier; (B) Schematic of an elastomer substrate with enhanced local crosslink density through UV patterning and a performance comparison with previous studies on rigid island applications. Reproduced with permission[103]. Copyright 2024, Wiley-VCH; (C) Schematic of TPV with varied modulus achieved by blending BPE and PLA (left), alongside a TEM micrograph of the TPV structure (right). Reproduced with permission[104]. Copyright 2015, The Royal Society of Chemistry. UV: Ultraviolet; TPV: thermoplastic vulcanizate; BPE: blending polyester elastomer; PLA: polylactide; TEM: transmission electron microscope.

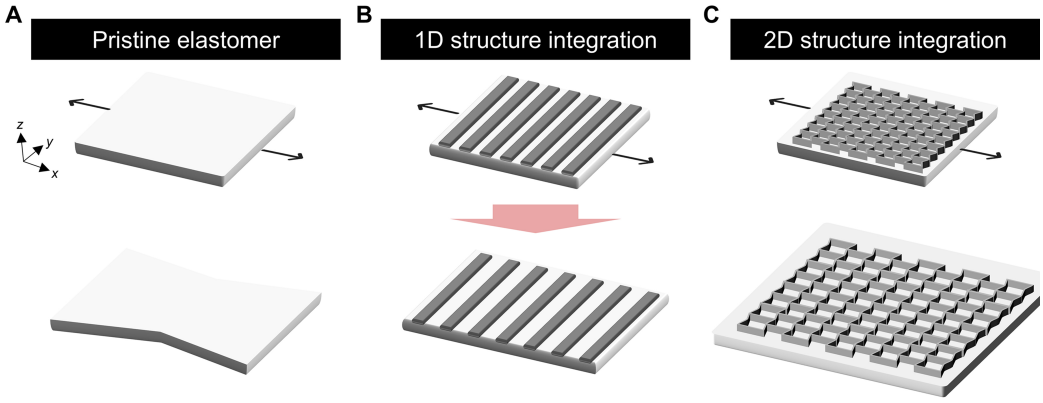

Figure 12. Schematic diagrams of elastomer films with integrated structures for strain control. (A) Pristine elastomer without structural integration; (B and C) Elastomer film with a (B) 1D structural frame and (B) 2D structural frame embedded within the elastomer matrices, respectively.

pattern, which was embedded into an elastomer matrix[109]. This resulted in a substrate with anisotropic deformation properties based on the alignment axis of the line pattern and the direction of stretching

[Figure 13A]. By coating the substrate with metal nanoparticles and allowing the distance and arrangement of these particles to vary with substrate tensile direction, the surface plasmonic effects produced a range of visible light wavelengths. This enabled the substrate to be used in plasmonic optical sensors capable of directional color tuning across a broad spectrum. Matsuda *et al.* conducted studies using a heterogeneous silicone substrate composed of PDMS and Ecoflex, employing a molding process to regulate spatial deformation distribution and overall mechanical deformation[111] [Figure 13B]. They fabricated dot-shaped PDMS pillars with high modulus and positioned them within the Ecoflex matrix to create rigid regions that suppress excessive deformation in pressure-sensitive areas. Additionally, they integrated porous silicone for pressure sensing and conductive silicone for strain sensing, enabling precise and independent measurements. This device maintains stable performance even under strains up to 50%, providing an effective solution for advanced sensing applications. Paik *et al.* developed a composite substrate capable of programmed anisotropic deformation by embedding 1D Cr patterns within an elastomer matrix and adjusting the width and spacing of these patterns[112] [Figure 13C]. The Cr line patterns were pre-fabricated on a silicon wafer using electron-beam lithography, followed by overcoating with a PDMS elastomer, curing, and subsequent detachment as a single layer. This composite substrate functions as a variable photomask, enabling diverse pattern light transmission depending on the designed deformation range, with adjustable deformation range and resolution based on the arrangement and width of the metal lines.

The 2D structure assembles frame elements capable of 2D deformation within the elastomer matrix, allowing for control of in-plane deformation through bi-directional or multi-directional expansion of the structure [Figure 12B]. Various types of planar-expanding structures, such as re-entrant, chiral, and Kagome grids, can be utilized in the form of frames, which are primarily fabricated using 3D printing methods[113] [Figure 14A]. Lee *et al.* developed a 2D re-entrant structure with a negative Poisson's ratio based on a honeycomb design and integrated it into an elastomer matrix[114]. By leveraging the competitive interaction between the expansion of the structure and the contraction of the matrix, they created a composite stretchable substrate with a negative Poisson's ratio, which was applied to a strain sensor, and this application improved the gauge factor by approximately 3.2 times compared to sensors without such structures [Figure 14B]. Additionally, Ha *et al.* conducted research on controlling the performance of resistive strain sensors by embedding a hard film with various integrated 2D patterns into the matrix[115] [Figure 14C]. The strain sensor was designed to concentrate surface deformation in specific areas, optimizing its performance to achieve both high sensitivity and stretchability while demonstrating long-term reliability.

Recently, innovative approaches have been exploring the integration of 3D structures into elastomer substrates to enable multi-dimensional deformation[117,118]. By employing intricate 3D designs, these methods aim to address the limitations of traditional planar electronic devices and investigate the potential of next-generation electronics with dynamic and adaptable 3D shapes. Such research goes beyond simple structural enhancements, seeking to fundamentally transform the interaction between substrates and devices in advanced applications, and drive significant technological advancements.

Rigid islands on elastomers for localized strain control

Previously, we concentrated on controlling the macroscopic deformation behavior of substrates by spatially adjusting the modulus patterns of elastomers. To expand the range of device applications, managing localized strain in areas where devices are positioned is essential; for this purpose, the rigid island pattern is commonly used. A rigid island involves placing island-shaped patterns with a higher modulus than the elastomer onto the substrate, thereby minimizing the deformation transmitted to the device under tensile strain[119-121]. Thus, positioning rigid devices on a stretchable substrate, with independent local deformation control, effectively mitigates damage or performance degradation in external deformation environments,

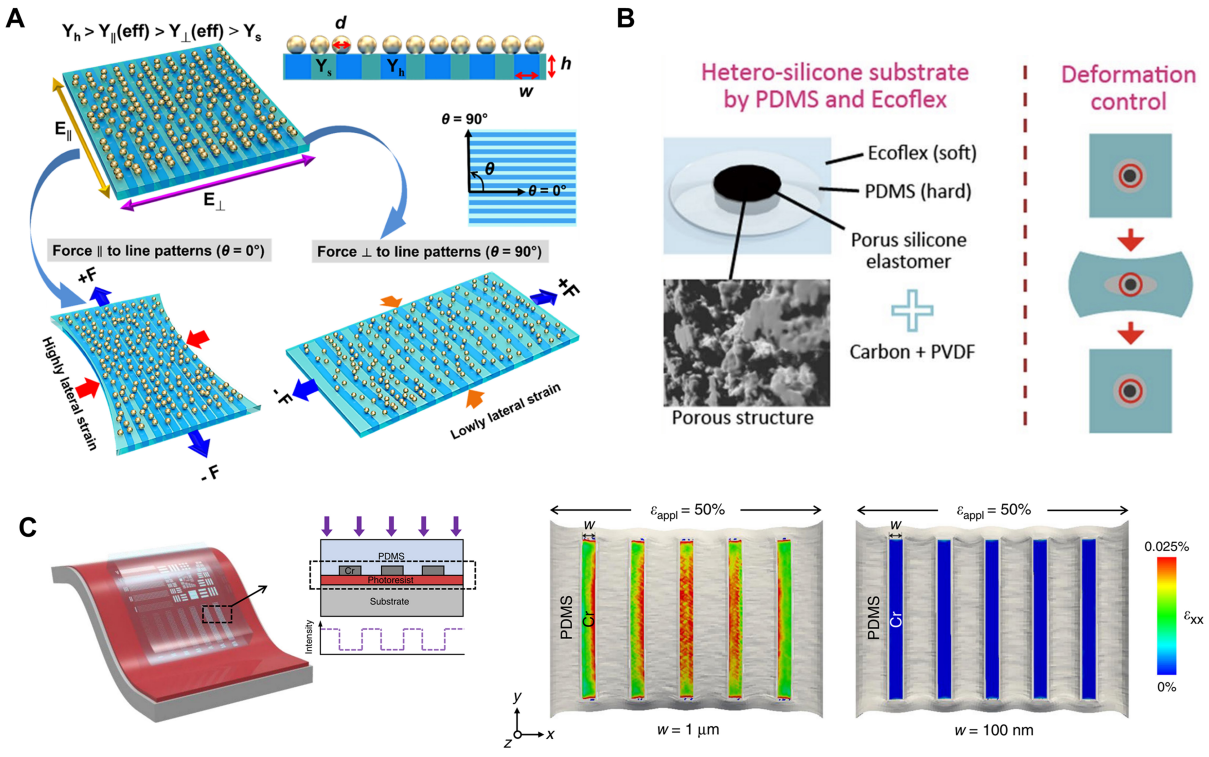

Figure 13. Elastomer matrix with integrated 1D structures. (A) Incorporation of 1D structures with high modulus line patterns into the elastomer matrix to regulate strain along the tensile axis, demonstrating an application in plasmonic meta-optical devices. Reproduced with permission[109]. Copyright 2024, American Chemical Society; (B) Integration of vertical pillar structures with high modulus into the elastomer matrix to prevent sensitivity loss in pressure sensors and to suppress excessive deformation in the sensing areas. Reproduced with permission[111]. Copyright 2020, Springer Nature; (C) Composite elastomeric substrate photomask embedded with 1D chromium line patterns, exhibiting diverse anisotropic deformation characteristics based on pattern and array conditions. Reproduced with permission[112]. Copyright 2020, Springer Nature.

necessitating specific design principles for successful implementation. First, the effective modulus of the rigid island should be appropriately designed by considering the target tensile deformation conditions, the device's deformation durability, and the moduli of both the device and the substrate. Generally, it should have a modulus significantly higher than that of the substrate and, ideally, higher than that of the device[119]. Second, robust bonding characteristics are required at each interface between the substrate, rigid island, and device, ensuring that the rigid island area where stress is concentrated can withstand repeated deformation within the target tensile range without delamination[120,121].

Rigid island patterns are typically formed by depositing heterogeneous materials onto the elastomer using mask patterns or by directly forming them through printing; sometimes, they are created by spatially patterning the crosslinking density of a homogeneous elastomer material[122,123]. Han *et al.* fabricated amorphous indium-gallium-zinc oxide (a-IGZO) thin-film transistors (TFTs) by layering a rigid island array made from PI material onto a stretchable polyurethane substrate[122] [Figure 15A]. Subsequently, they applied an additional 3 μm thick organic passivation layer on top of the TFTs to further enhance mechanical durability. As a result, the produced stretchable TFTs maintained their electrical performance even after 10,000 stretching cycles at a strain rate of 30%. Kang *et al.* successfully implemented high-stretchability inorganic transistors by embedding rigid island transistors within an elastomer substrate[123] [Figure 15B]. Among various combinations of rigid island and stretchable elastomer materials, the pairing of polyepoxy acrylate (PEA) and polyurethane acrylate (PUA) enabled strong covalent bonding, achieving robust

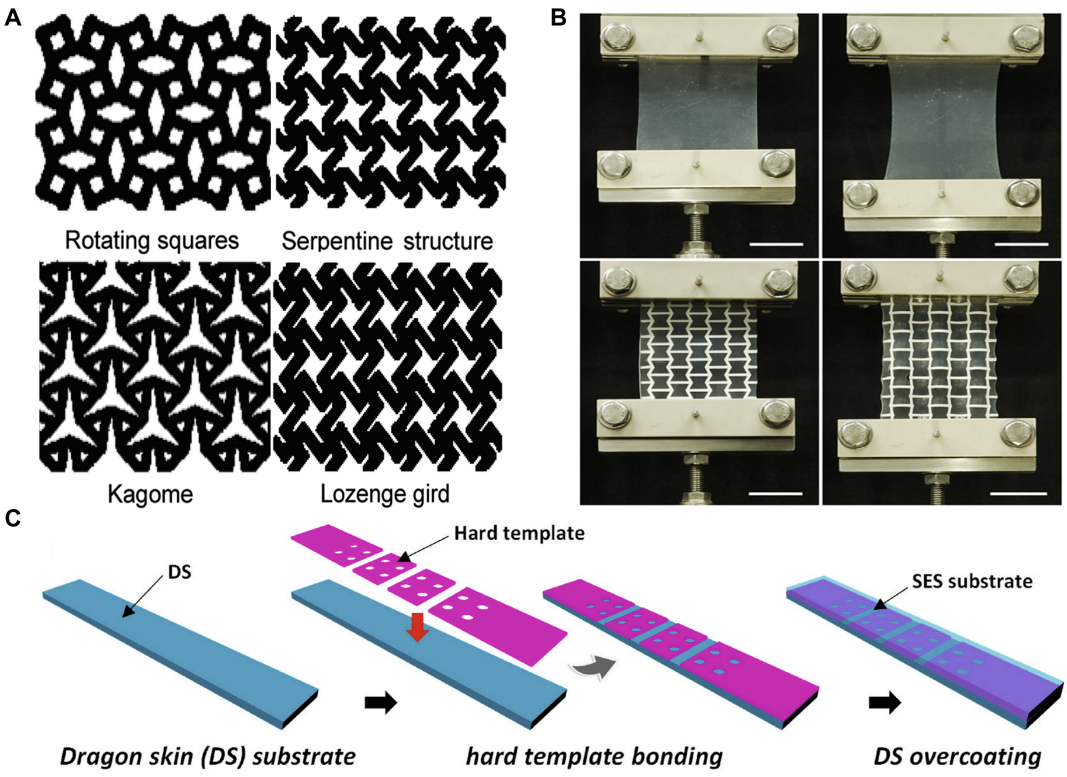

Figure 14. Elastomer matrix incorporating 2D structures. (A) Examples of various designs of 2D mechanical metastructures that can be integrated into the elastomer matrix for strain control. Reproduced with permission[113]. Copyright 2023, Wiley-VCH; (B) Comparison of vertical contraction deformation during stretching of the elastomer film with and without the 2D strain control structures. Reproduced with permission[114]. Copyright 2019, Elsevier; (C) Schematic of the fabrication process for an elastomer film with embedded structures designed for tailored deformation in different regions. Reproduced with permission[115]. Copyright 2022, Elsevier.

adhesion at the interface. Consequently, the transistors maintained their functionality under strains exceeding 50% and retained stable electrical performance even after 10,000 repeated stretching cycles.

As previously discussed, integrating rigid islands with careful consideration of the elastic moduli of both the substrate and the device can significantly enhance the mechanical reliability of devices in stretchable environments, and recent approaches have focused on precisely designing rigid islands to exhibit a composite modulus along the thickness direction. This precise design allows for more effective control of localized deformations, minimizing stress at each interface and redistributing it to surrounding areas, thereby reducing stress concentrations at interfaces and further improving the device's mechanical reliability and durability[124-126]. Kim *et al.* developed rigid islands in a multilayer polymer film (MLPF) with modulus engineering to enhance the mechanical durability and reliability of stretchable electronic devices[125]. This MLPF was produced using a layered CVD process, with precise adjustments of each layer's modulus between the substrate and the islands to achieve composite modulus along the thickness direction, ensuring the mechanical reliability of devices formed on the islands under tensile strain [Figure 15C]. By restricting the deformation of the rigid islands to below 1% across the full strain range, this system was successfully integrated with a-IGZO TFTs and maintained stable performance even after 100,000 cycles at 30% strain. Additionally, Sun *et al.* introduced a composite stretchable substrate design featuring a gradient modulus zone in the thickness direction within regions where rigid islands would be formed, aiming to enhance induced stress management during tensile deformation[126] [Figure 15D]. They soaked the PDMS substrate in a methacrylic acid (MAA) monomer solution, allowing it to absorb curing agents, and then

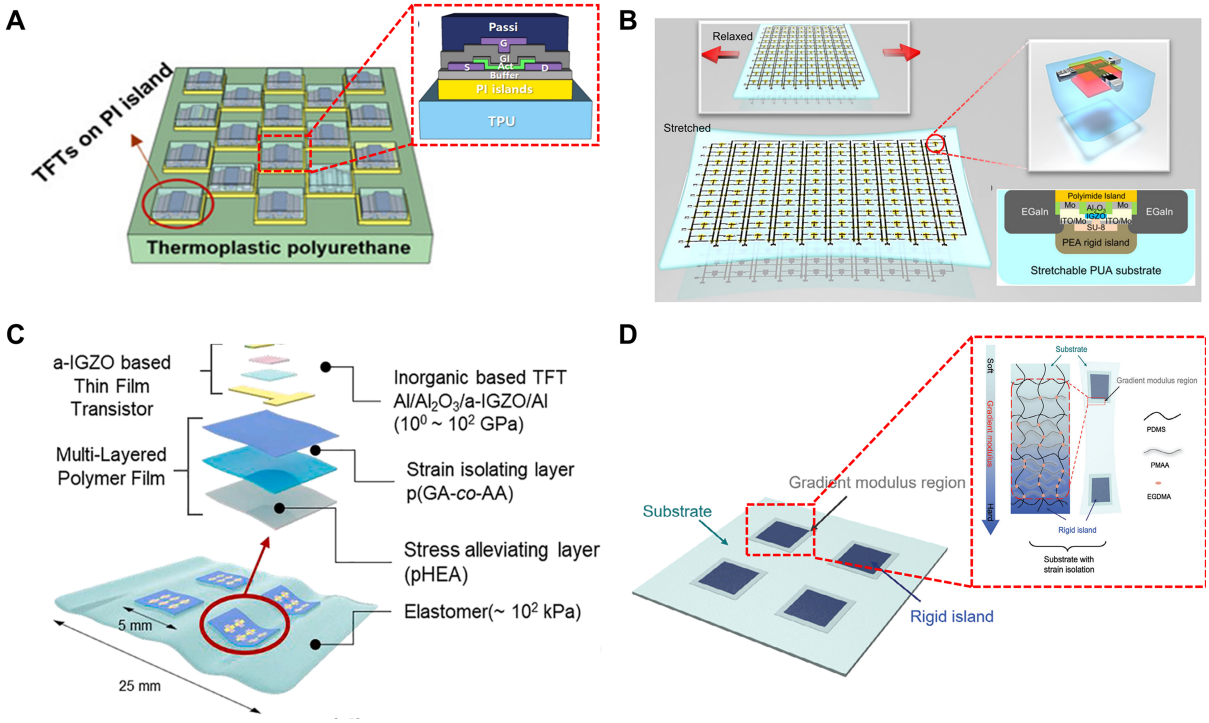

Figure 15. (A) PI-based rigid island array assembled on a TPU substrate for a stretchable TFT. Reproduced with permission[122]. Copyright 2021, American Chemical Society; (B) Stretchable inorganic transistor with a PEA elastomer of relatively higher modulus embedded as a rigid island array within a PUA substrate, exhibiting excellent adhesion at the PUA-PEA interface. Reproduced with permission[123]. Copyright 2024, Springer Nature; (C) MLPF with varying moduli introduced as rigid islands on a stretchable substrate, enabling a stretchable inorganic TFT. Reproduced with permission[125]. Copyright 2022, Elsevier; (D) Stretchable substrate with an array region designed with a modulus gradient along the thickness direction to optimize stress distribution during tensile deformation. Reproduced with permission[126]. Copyright 2024, Wiley-VCH. PI: Polyimide; TPU: thermoplastic polyurethane; TFT: thin-film transistor; PEA: polyepoxy acrylate; PUA: polyurethane acrylate; MLPF: multilayer polymer film.

used patterned UV exposure to form a gradient modulus based on the UV penetration characteristics through PDMS in the thickness direction. On this engineered composite stretchable substrate, they transferred a serpentine electrode and attached a light-emitting diode (LED), verifying mechanical stability and stress concentration relief through repeated tensile and deformation tests.

DISPLAY APPLICATIONS OF STRAIN-ENGINEERED STRETCHABLE SUBSTRATES

This section presents examples of how strain-engineered stretchable substrates are being applied in various free-form displays. Light-emitting devices include micro-LEDs, quantum dot LEDs (QD-LEDs), and OLEDs, which are utilized in combination with different types of stretchable substrates depending on target application, working environments, and deformation patterns[127-131]. Micro-LEDs are typically fabricated on mother substrates and subsequently transferred onto stretchable substrates using transfer printing methods[127,128]. For QD-LEDs and OLEDs, beginning with deposition processes, recent advancements in materials and techniques have introduced low-temperature inkjet printing as a method for fabricating light-emitting parts[129,130]. Among the proposed strain-engineered stretchable substrates, structured plastic substrates offer relatively superior thermal and mechanical stability compared to elastomers. As ongoing research enhances thermal stability while maintaining substrate transparency, it has become possible to fabricate devices using existing display process lines[132,133]. This allows for facile integration with existing display materials and processes, accelerating product commercialization. However, issues such as trade-offs between stretchability and resolution and limited degrees of deformation freedom exist.

Conversely, elastomer substrates with inherent stretchability offer substantial advantages over the previously discussed structured plastic substrates from a free-form display standpoint[134,135]. These benefits include enabling high-resolution applications and maintaining stable deformation in environments with dynamic and multiple deformations. To fully leverage these benefits, it is desirable for the light-emitting parts formed on the substrate to also possess stretchability. Proposed methods include forming wrinkle structures on the elastomer surface to impart structural stretchability to the light-emitting parts[136,137] and developing new materials to construct light-emitting parts with inherent stretchability[138,139]. In summary, the ideal combination of light-emitting components and strain-engineered substrates is chosen based on the specific demands of free-form displays, with Table 1 presenting the properties of each stretchable substrate and key considerations for their application in free-form displays.

Display applications on structured plastic substrates

As previously mentioned, structured plastic substrates have been widely studied in early stretchable display research due to their structural simplicity and high manufacturing efficiency. This approach allowed companies to maintain existing display processes while avoiding complex additional steps, leading to extensive adoption and the presentation of prototype products at numerous exhibitions. For instance, Jeong *et al*. employed a plastic composite substrate with precisely controlled short wrinkle wavelengths to mitigate pixel distortion caused by traditional wrinkle structures used for stretchability in flexible substrates and to reduce spatial loss in displays[140]. In this study, a thin PI-based plastic composite substrate was utilized to fabricate a thin-film LED (TFLED). Compared to conventional macroscopic wrinkle structures, the short-wavelength wrinkle structure with a 20 μm wavelength significantly reduced visible image distortion, demonstrating excellent visual quality and potential for application in high-resolution free-form displays and wearable devices [Figure 16A]. Kim *et al*. developed a wearable QD-LED for stretchable optoelectronic applications by incorporating a wrinkle structure on a polyethylene naphthalate (PEN) substrate, which offers superior thermal stability compared to PET[141]. After forming the device on the PEN substrate, it was transferred to a pre-stretched elastomer substrate, creating a wrinkle structure that allowed the device to operate stably under stretch conditions of up to 70% [Figure 16B]. Furthermore, the elastomer beneath the plastic layer enabled easy attachment to the skin, demonstrating potential for use in various wearable electronic applications. Additionally, Yin *et al*. developed an energy self-powered stretchable display system by transferring a stretchable OLED and a stretchable polymer solar cell onto a plastic substrate, then forming a periodic buckling structure[142]. Using a stencil-based deposition method, they created alternating adhesive and non-adhesive regions on the substrate, attaching devices to a pre-stretched substrate and releasing it to achieve the buckled display system. This configuration achieved a maximum stretchability of 100% and consistently maintained performance even after more than 20,000 cycles of stretching and recovery [Figure 16C].

Kirigami-structured flexible plastic substrates are widely used in stretchable display applications, particularly for controlling in-plane deformations such as Poisson's ratio. Jang *et al.* introduced research that combines an auxetic meta-structure with a Kirigami pattern to improve image distortion in stretchable displays[143]. The auxetic metamaterial, which has a negative Poisson's ratio, expands laterally when stretched axially, allowing for stable display operation even under 24.5% stretch conditions. The designed Poisson's ratio of -1 helps maintain the original image after stretching, and this design allows for uniform deformation when attached to curved surfaces [Figure 17A], making it suitable for skin-attached phototherapy devices or displays that can be affixed to non-spherical surfaces. Deng *et al.* implemented a display device using a PEN substrate with a rotating square Kirigami pattern, designed for applications in curved tensile environments[144]. The integration of the driving flexible printed circuit board (FPCB) and LED elements on the PEN substrate provided up to 57% bidirectional stretchability, and the device demonstrated stable operation even on complex 3D curved surfaces such as cylindrical, spherical, and

Table 1. Characteristics of strain-engineered stretchable substrates and their advantages and limitations in free-form display applications

Substrate type	Plastic film		Elastomers		
	Buckling	**Kirigami**	**Network aligning**	**Crosslinking control**	**Structure assembling**
Stretchable characteristic	Vertical compensation	Void compensation	Intrinsic stretchability		
Representative manufacturing	Pre-stretching Mold transfer	Laser cutting Etching	Rubbing Post-stretching	Thermal patterning Light patterning	Evaporation Direct printing
Advantages in free-form display	Facilitates application in conventional display materials and process lines		Suitable for high-resolution and multi-form applications		
Limitations in free-form display	Trade-off between stretchability and resolution/reliability		Requirement for developing new stretchable materials to overcome process limitations		

saddle-shaped surfaces [Figure 17B]. To address the issue of resolution degradation in stretchable displays due to stretching, Lee *et al.* proposed a stretchable OLED device that maintains a high fill factor even after stretching by exposing hidden active areas (HAA) during Kirigami deformation[145]. They attached an ultrathin OLED to a 3D Kirigami-structured substrate, allowing the HAA to become visible during biaxial stretching, thereby preserving resolution and quality [Figure 17C]. This approach to compensating for resolution loss effectively demonstrates the potential of the proposed method in maximizing the capabilities of stretchable OLEDs.

Display applications on intrinsically stretchable elastomer substrates

Elastomers, which enable omnidirectional stretchability without compensatory structures, are considered highly promising as substrates for free-form displays, with active research underway to enhance their thermal stability and mechanical properties for effective display component integration[146,147]. Additionally, as described in previous sections, efforts are also underway to expand the potential applications of free-form displays through macroscopic and localized strain-engineering. Oh *et al.* developed a method to improve image distortion in stretchable displays caused by the Poisson effect of elastomers by embedding various 1D line patterns into an elastomer matrix[148]. This approach effectively suppresses the contraction due to the Poisson effect during stretching, resulting in a composite elastomer substrate with a near-zero Poisson's ratio [Figure 18A]. To minimize unwanted deformation of the substrate, the researchers meticulously designed the modulus, dimensions, and shape of the line patterns and used transparent materials to ensure excellent optical transparency of the composite substrate. To enable diverse deformation applications while maintaining a zero Poisson's ratio for the substrate, Choi *et al.* developed a meta-elastomer substrate with a bidirectional zero Poisson's ratio by embedding structurally designed honeycomb-shaped soft mechanical metamaterials into an elastomer matrix[149]. This substrate utilized soft materials for the frame, allowing for independent tensile deformation along different axes [Figure 18B]. By optimizing the dimensions of the structures and the modulus mismatch between the substrate and the matrix, they effectively controlled the anisotropic Poisson's ratio. The LED pixel arrays formed on this zero Poisson's ratio meta-elastomer substrate demonstrated linear and predictable deformation during stretching, indicating significant potential for applications in stretchable displays that require various in-plane shape deformations. Lee *et al.* proposed a stretchable display design where a structured plastic film with a negative Poisson's ratio is embedded within an elastomer matrix, allowing the display image to expand biaxially under tension[150] [Figure 18C]. The substrate integrates a rigid glass-fiber reinforced PDMS (GFRPDMS) region with a soft elastomer matrix, optimizing the elastic modulus difference between the stiff and stretchable areas to maintain stability under multi-directional stretching and achieve consistent strain control and performance stability, even with repeated deformation. Using this substrate, they developed a stretchable display incorporating a high-resolution micro-LED array and liquid metal electrodes, achieving up to 25% stretch without image degradation.

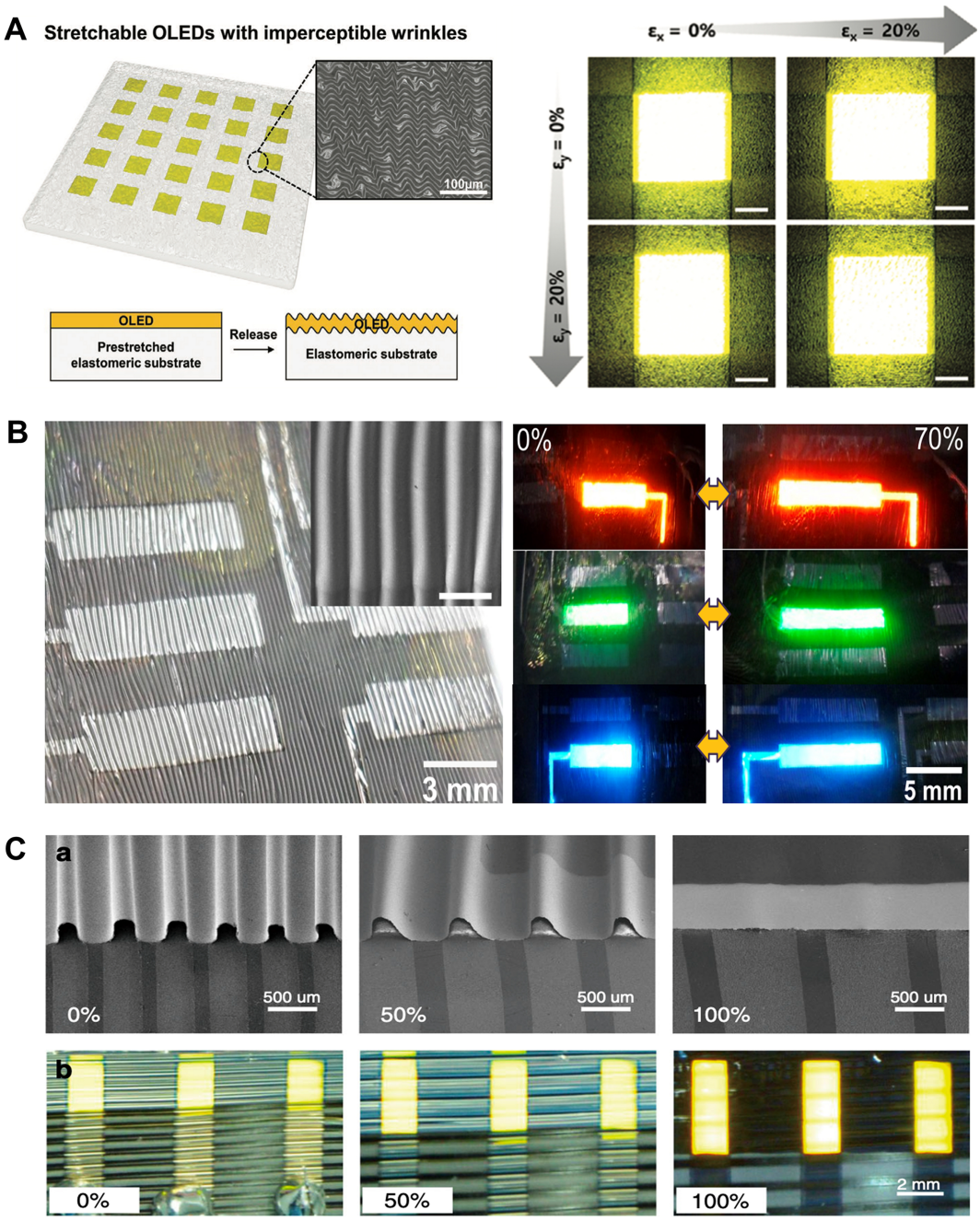

Figure 16. Display applications utilizing buckling-structured flexible films. (A) Schematic diagram of the concept for an OLED with an invisible wrinkle structure, surface SEM and light luminescence images before and after stretching, based on the dimensions of the wrinkle structure introduced on the TFLED device substrate. Reproduced with permission[140]. Copyright 2020, Wiley-VCH; (B) Photographs of a pre-strained, wavy QD-LED. The inset shows a magnified SEM image (scale bar, 300 μm), buckling structure of the flexible substrate under tension and the R/G/B luminescence characteristics of stretchable QD-LEDs under stretching and bending conditions. Reproduced with permission[141]. Copyright 2017, American Chemical Society; (C) SEM images and photographs of SOLEDs captured in a dark environment under tensile strains of 0%, 50%, and 100%. Reproduced with permission[142]. Copyright 2018, Nature Publishing Group. OLED: Organic light-emitting diode; SEM: scanning electron microscope; TFLED: thin-film light-emitting diode; QD-LED: quantum dot light-emitting diode; SOLEDs: stretchable organic light-emitting devices.

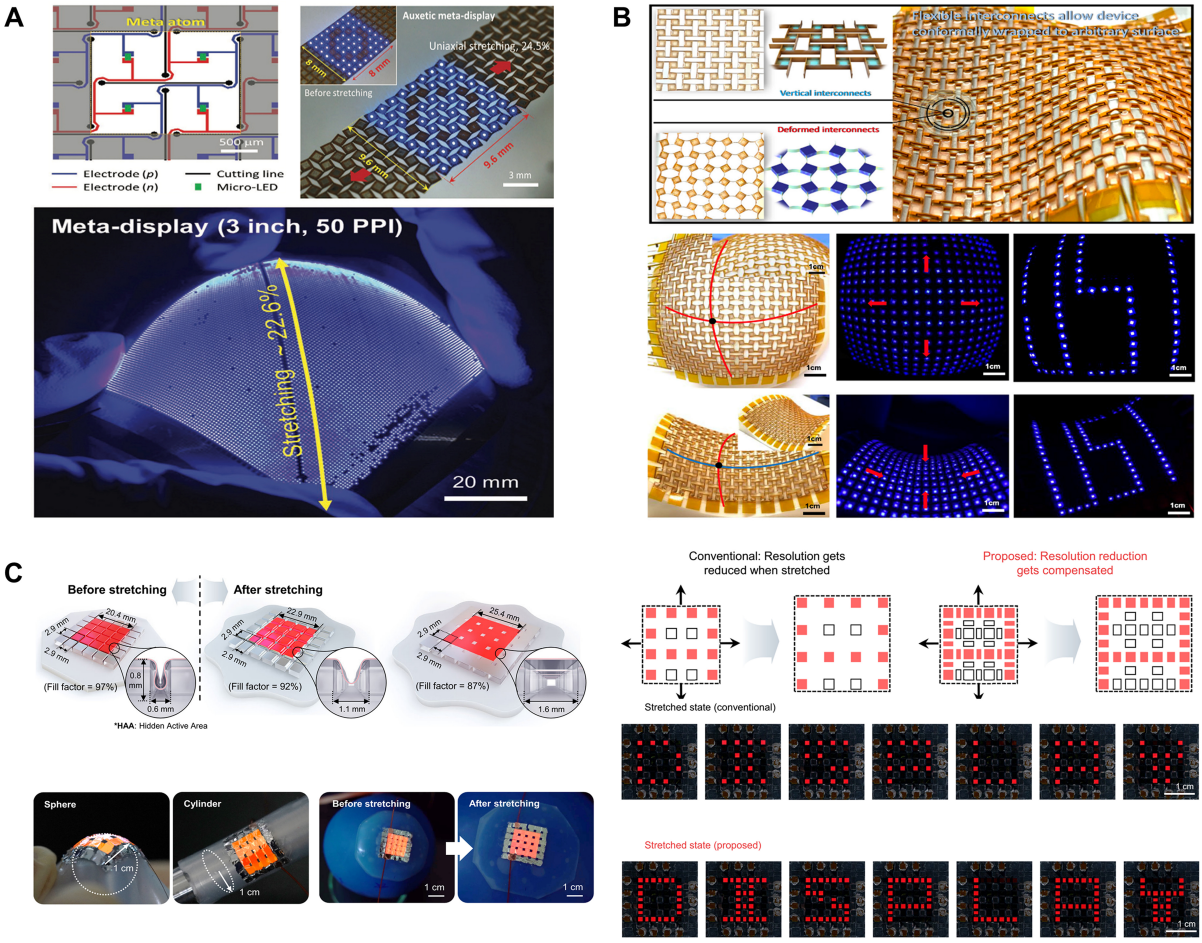

Figure 17. Examples of flexible displays utilizing Kirigami patterns. (A) A meta-display designed with a Kirigami pattern that has a Poisson's ratio of -1 to address image distortion caused by the Poisson effect, along with examples of luminescence on curved surfaces. Reproduced with permission[143]. Copyright 2022, Wiley-VCH; (B) A stretchable display with rotational Kirigami patterns demonstrating stable luminescence properties on various curved surfaces, including cylindrical, spherical, and saddle shapes. Reproduced with permission[144]. Copyright 2024, Springer Nature; (C) A stretchable OLED that reveals HAA through Kirigami deformation, maintaining high resolution even after stretching. Reproduced with permission[145]. Copyright 2024, Springer Nature. OLED: Organic light-emitting diode; HAA: hidden active areas.

In addition to macro-scale planar deformations, controlling localized deformation where the device is formed is crucial for practical applications. Strategies for engineering the modulus in the vertical direction are commonly employed to achieve this. To address these needs, Kim *et al.* proposed a hybrid platform with a dual-layer structure incorporating a stress-relief layer and a rigid island array on an elastomer substrate to prevent performance degradation in stretchable OLEDs under repeated stretching and restoring conditions[151]. This platform effectively manages localized strain applied to the device formed on the islands by engineering the modulus in the thickness direction. As a result, the device can operate within environments stretched up to 140% while maintaining excellent electromechanical and optical performance, demonstrating outstanding stability even after repeated deformations [Figure 18D].

CONCLUSION AND OUTLOOK

This review provides a comprehensive summary of research trends on strain-engineered stretchable substrates for free-form display applications. It primarily proposes design strategies utilizing strain-compensation structures and modulus patterning, focusing on structured plastic films and intrinsically

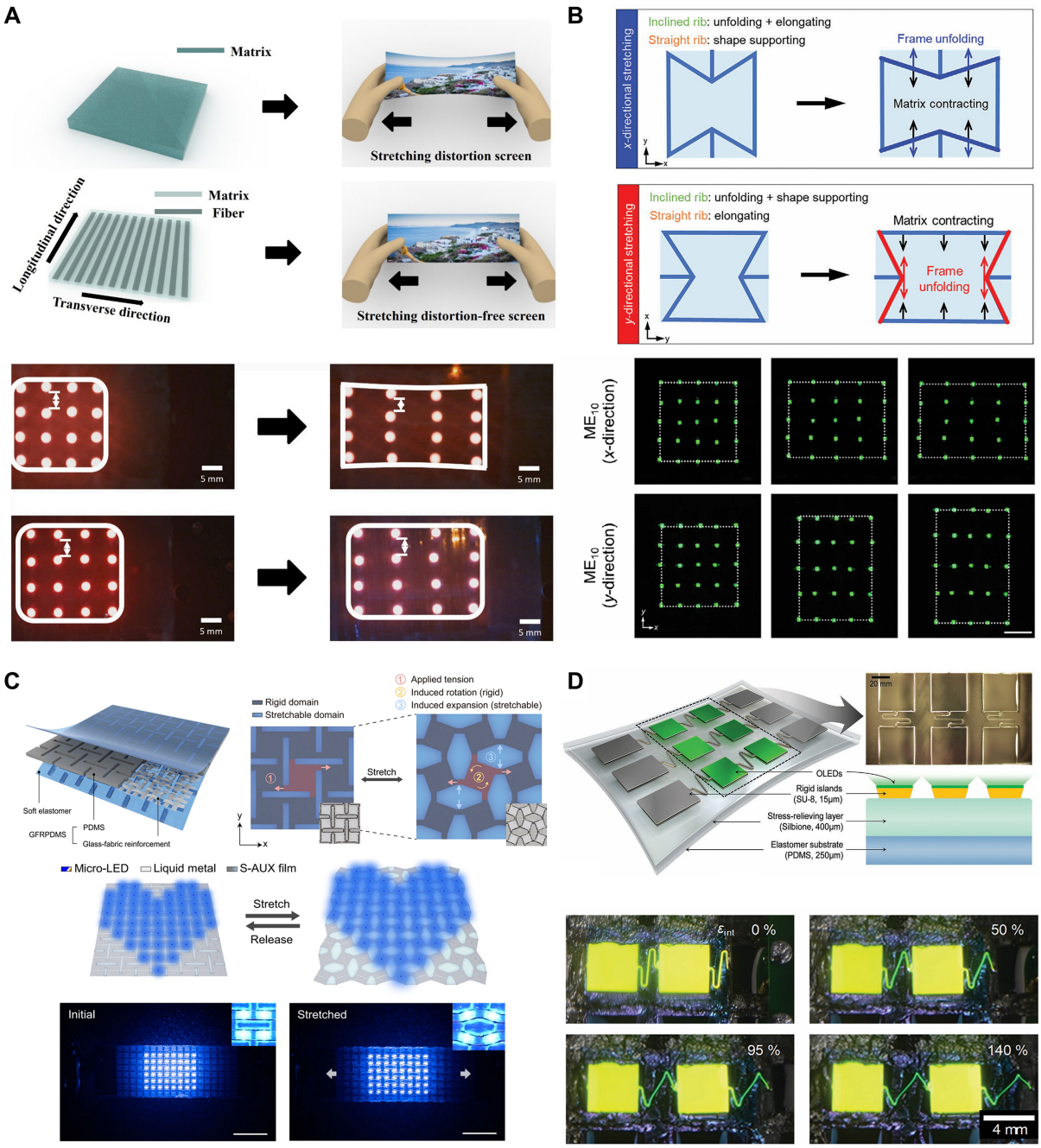

Figure 18. (A) Integration of 1D line patterns with various shapes and sizes into an elastomer matrix to adjust the substrate's Poisson's ratio to nearly zero and improve image distortion in stretchable displays. Reproduced with permission[148]. Copyright 2024, Elsevier; (B) Incorporation of a soft mechanical meta-material frame capable of self-deformation into an elastomer matrix to achieve a zero Poisson's ratio in the biaxial direction, and examples of linear and predictable display pixel movement under biaxial tensile deformation. Reproduced with permission[149]. Copyright 2024, Wiley-VCH; (C) Stretchable displays featuring a Kirigami structure based on high-modulus GFRPDMS embedded within an elastomer matrix, which exhibits a negative Poisson's ratio, enabling biaxial expansion deformation without image distortion under tensile strain. Reproduced with permission[150]. Copyright 2024, Springer Nature; (D) A hybrid platform with a dual-layer structure, incorporating a stress-relief layer and a rigid island array on an elastomer substrate, engineered with modulus variations in the thickness direction to prevent performance degradation in stretchable OLEDs. Reproduced with permission[151]. Copyright 2020, Wiley-VCH. GFRPDMS: Glass-fiber reinforced polydimethylsiloxane; OLEDs: organic light-emitting diodes.

stretchable elastomer substrates. The review also explores how to maximize these approaches in various environments through careful selection of substrate materials and strain designs tailored to specific applications. Initially, promising buckling and Kirigami structures are examined, with an analysis of the tensile deformation characteristics of structured plastic substrates under different design conditions. Specifically, the review addresses deformation characteristics of buckling structures based on shape, size, and isotropy, and explores Kirigami structures by systematically analyzing the effects of cut pattern shape, size, and combinations, providing design guidelines for plastic film substrates across diverse applications. Furthermore, the review discusses strategies for using intrinsically stretchable elastomer materials, particularly approaches to spatially pattern moduli to minimize unwanted deformations in pristine elastomers. This section also discusses techniques such as inducing network alignment or crosslink patterning in homogeneous elastomers through external stimuli, along with utilizing compensatory effects in heterogeneous structures to achieve multi-dimensional control over both local and global deformations.

In free-form display applications, electrodes, driving components, and light-emitting elements are layered onto strain-engineered stretchable substrates, where the choice of substrate material and strain-engineering strategy should be carefully tailored to factors such as the type of light-emitting element, processing temperature, target resolution, and deformation behaviors. For example, while structured plastic substrates have limited effective areas for device formation, they offer greater heat resistance and mechanical stability than elastomers, ensuring compatibility with existing display manufacturing processes. Due to these properties, plastic substrates can be quickly commercialized in low-resolution applications such as simple signal displays or automotive screens. Conversely, elastomers provide intrinsic stretchability without additional structures, offering a large effective area, making them suitable for high-resolution displays. However, their low thermal stability restricts processing temperatures, necessitating further research to develop materials with enhanced thermal stability or reduced processing temperatures[152].

To successfully develop displays that support various form factors, ensuring compatibility between the strain-engineered stretchable substrates proposed in this review and key display components is essential. Additionally, for commercialization, hardware or software solutions are required to compensate for resolution loss and enable seamless content transitions due to form-factor changes[153]. Ideally, for high-resolution, multi-dimensional displays, most display components, including the substrate, should possess inherent stretchability. Free-form display technology is currently considered to be between the introductory and growth stages, with intensive research underway on materials, manufacturing processes, and software-based image compensation to develop optimized protocols for specific applications. However, challenges remain regarding resolution, deformation characteristics, and mechanical reliability, necessitating ongoing research and innovation to address these issues. The strain-engineering design strategies proposed in this review are expected to help overcome some of these limitations, supporting the reliable operation of free-form displays across various deformation conditions. This advancement could play a crucial role in expanding the application potential of free-form displays from static shape transformations to real-time dynamic deformations.

DECLARATIONS

Authors' contributions

Conceptualization and supervision: Choi JC, Chung S
Prepared the figures and wrote the article: Lee DW, Park DH, Choi JC

Availability of data and materials

Not applicable.

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Conflicts of interest

Chung S is the Guest Editor of the Special Issue, while the other authors have declared that they have no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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