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Topical Review

Advancements in transfer printing techniques for flexible electronics: adjusting interfaces and promoting versatility

Zijian Chen[1,6], **Chi Zhang**[2,6] **and Zijian Zheng**[1,2,3,4,5,*]

[1] School of Fashion and Textiles, The Hong Kong Polytechnic University, Hong Kong Special Administrative Region of China, People's Republic of China
[2] Department of Applied Biology and Chemical Technology, The Hong Kong Polytechnic University, Hong Kong Special Administrative Region of China, People's Republic of China
[3] State Key Laboratory for Ultra-Precision Machining Technology, The Hong Kong Polytechnic University, Hong Kong Special Administrative Region of China, People's Republic of China
[4] Research Institute for Intelligent Wearable Systems, The Hong Kong Polytechnic University, Hong Kong Special Administrative Region of China, People's Republic of China
[5] Research Institute for Smart Energy, The Hong Kong Polytechnic University, Hong Kong Special Administrative Region of China, People's Republic of China

E-mail: tczzheng@polyu.edu.hk

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Abstract

The burgeoning interest in flexible electronics necessitates the creation of patterning technology specifically tailored for flexible substrates and complex surface morphologies. Among a variety of patterning techniques, transfer printing emerges as one of the most efficient, cost-effective, and scalable methods. It boasts the ability for high-throughput fabrication of 0–3D micro- and nano-structures on flexible substrates, working in tandem with traditional lithography methods. This review highlights the critical issue of transfer printing: the flawless transfer of devices during the pick-up and printing process. We encapsulate recent advancements in numerous transfer printing techniques, with a particular emphasis on strategies to control adhesion forces at the substrate/device/stamp interfaces. These strategies are employed to meet the requirements of competing fractures for successful pick-up and print processes. The mechanism, advantages, disadvantages, and typical applications of each transfer printing technique will be thoroughly discussed. The conclusion section provides design guidelines and probes potential directions for future advancements.

[6] These authors contribute equally to the work.
[*] Author to whom any correspondence should be addressed.

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1. Introduction

The burgeoning trend of human-device integration, encompassing personalized health monitoring and medication, biomechatronic prosthetics, and human augmentation technologies, necessitates a significant demand for electronic devices that are air and moisture permeable, surface conformable and mechanically deformable. These devices are anticipated to be as comfortable as clothing, catering to the emerging demands of the Internet of Things (IoT), which facilitates real-time human interaction with the physical world [1–4]. Flexible electronics technology, characterized by its softness, light weight, transparency, and portability, holds the potential to transcend the intrinsic constraints of conventional silicon-based optoelectronics as we move beyond the Moore's era. Poised at the forefront of multidisciplinary convergence, this technology is set to drive innovation across information technology, healthcare, aerospace, and advanced energy sectors, among others [5, 6]. However, manufacturing flexible electronics remains a formidable task because the key component of contemporary electronic devices, integrated circuits (ICs), are normally fabricated by conventional patterning methods, such as the most well-known photolithography and electron beam lithography [7, 8], which are typically designed for planar and rigid substrates. As the fabrication resolution is continually refined to several nanometers [9], the feature size of the devices becomes commensurate with the amplitude of surface roughness (nano to micrometers) [10]. Whereas, most flexible substrates are challenging to wet, incompatible with strong solvents, and thermally unstable, leading to significant challenges in the patterning process for wearable electronics [11–15].

Numerous efforts have been devoted to developing various advanced patterning techniques to address these issues, such as printing techniques and soft lithography [16–21]. Transfer printing (TP) and its derivative patterning methods, as one of the most promising representatives, have significantly contributed to the fabrication of various photonic/electronic devices. Rogers' group first proposed a TP process in 2006 [22]. As shown in figure 1, the method involves a 'pick-up' process and a subsequent 'print' process which is realized by utilizing an elastomer stamp [e.g. polydimethylsiloxane (PDMS)]. The pre-fabricated devices or components are peeled off from the original donor substrate by the elastomer stamp and serve as the 'ink' part during the following print process. The devices are transferred to a receiver substrate by conformal contacting of the 'inked' stamp and the target substrates through adhesion force dominated by van der Waals forces [23, 24]. The success of this process is attributed to the viscoelastic behavior of the stamp, where the adhesion force between the stamp and the devices is proportionate to the peeling speed. Applying a large enough peeling speed during the peel-off process will supply a strong adhesion force to take the device away from the donor substrate while, on the contrary, an extremely low peeling speed will decrease the adhesion force and leave the devices attached to the receiver substrate [22, 25, 26]. Leveraging powerful methodologies and theoretical frameworks, such as Atomic Force Microscopy (AFM) and the Johnson–Kendall–Roberts theory, researchers have conducted thorough investigations into the adhesion and friction properties of elastomers. These studies, which delve into the relationships between molecular structures and functional groups, have significantly advanced our understanding and application of PDMS elastic stamps throughout the past several decades [27, 28]. For example, reverse offset printing techniques have made substantial contributions to the field of TP. By employing semi-dried ink layers, these methods shift the underlying mechanism from one dominated by wetting to one governed by adhesion and friction. This strategic adjustment significantly mitigates the issues associated with ink diffusion, thereby enhancing the resolution of the printed patterns [29, 30].

TP offers several advantages over its counterparts. Firstly, it is highly compatible with traditional photolithography methods, which facilitates the retrofitting of existing production processes. On the one hand, the resolution is superior to other printing technologies such as screen printing [25, 31], inkjet printing [32–35], and so on. On the other hand, the mass production capability outperforms many patterning methods like dip-pen lithography [36–39]. Secondly, TP inherits the advantages of lithography while expanding the categories of receiver substrates with flexibility and stretchability. The types of ink materials are also varied, including metals [40, 41], polymers [42–44], organic or inorganic materials [33, 45–47], and others. Thirdly, the use of the elastomer stamp eliminates the possibility of chemical corrosion and contamination, expanding the category of acceptable flexible receiver substrates [11]. Furthermore, the transfer process can be conducted under mild biocompatible conditions cost-effectively, expanding the applicable scenarios to biomedical areas [48–50]. The ability of multilayer TP with high precision has also been well developed over the years [51–53].

According to theoretical calculation and experiments results, the success of TP relies on the competing relationship between the energy release rate (G) of the interfaces. This relationship is contingent on the regulation of adhesion force F between the three interfaces: (1) donor substrate-devices (F_1), (2) stamp-devices (F_2), and (3) devices-receiver substrate (F_3) [54]. The pick-up and print processes require the $F_1 < F_2$ and $F_{2'} < F_3$, respectively [55–57]. It is noteworthy that F_2 may differ from $F_{2'}$, as they correspond to the stamp-device interface under varying conditions, such as loading rate and temperature.

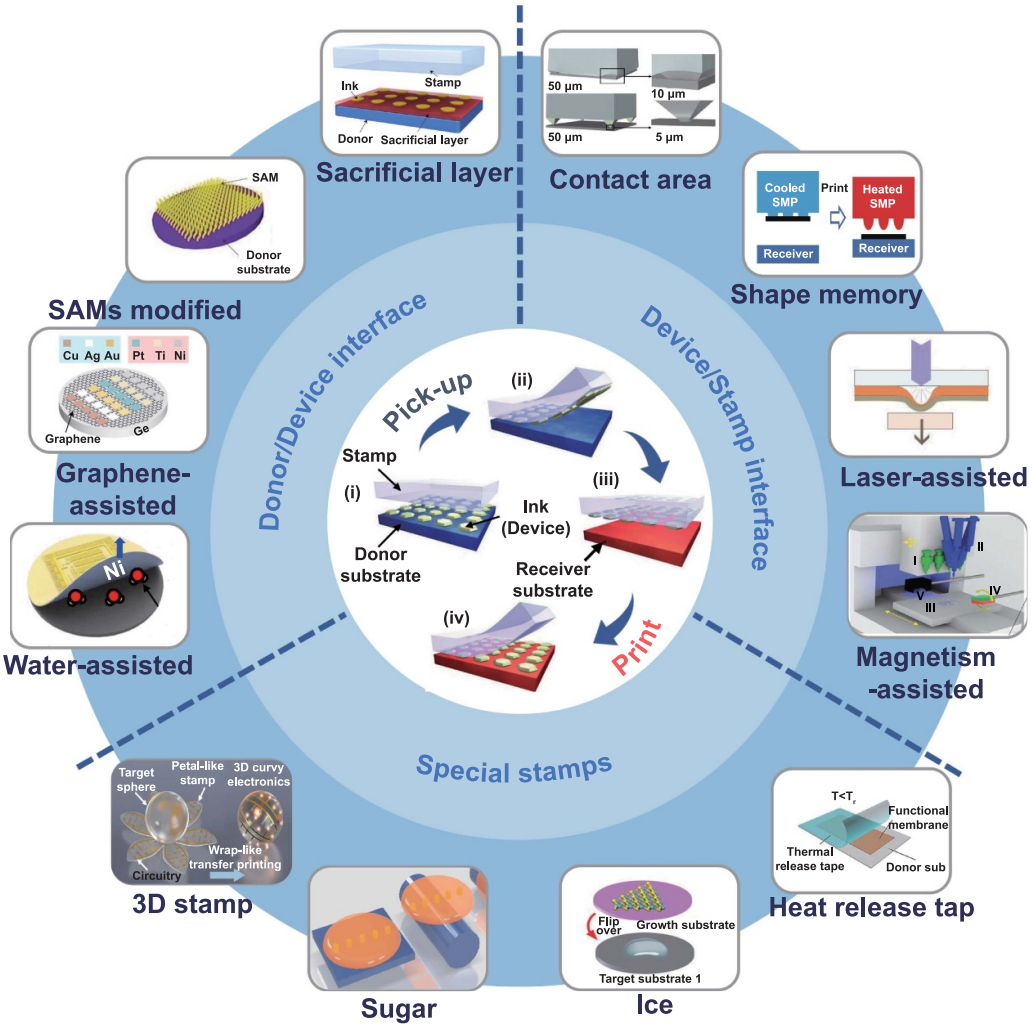

Figure 1. Summary of the mechanism of TP and technical core to promote the versatility for high-yield fabrication of flexible electronics. The TP process could be divided into four steps: (i) laminating the stamp on the donor substrate with pre-fabricated devices, (ii) picking up the devices from the donor substrate with high peeling speed, (iii) placing the stamp/devices layer onto the surface of the receiver substrate, (iv) removing the stamp with low peeling speed and leaving the devices on to the receiver substrate. Adapted from [22], with permission from Springer Nature. To improve the success rate during the transfer process, various modifications could be applied to the donor/device interface, the device/stamp interface and the special design for the stamp. For the donor/device interface, water. Reproduced with permission from [58], © 2018. Published under the PNAS license. Graphene. Reproduced from [59], with permission from Springer Nature. SAMs. Reproduced from [60], with permission from Springer Nature, and sacrificial layer. Reproduced from [61]. CC BY 4.0, were utilized. For the device/stamp interface, change of contact area by elastomer. Reproduced from [62]. © 2010. Published under the PNAS license, and shape memory materials. Reprinted from [63], © 2021 The Author(s). Published by Elsevier Ltd on behalf of The Chinese Society of Theoretical and Applied Mechanics, laser. Reprinted from [64], Copyright © 2012 Elsevier B.V. All rights reserved, and magnetism. Reproduced from [65]. CC BY 4.0., were utilized. For the special stamp design, heat release tap. Reproduced from [66]. CC BY 4.0, ice. [67] John Wiley & Sons. © 2023 Wiley-VCH GmbH, sugar. From [68]. Reprinted with permission from AAAS, and 3D stamp. Reproduced from [69]. CC BY 4.0, were utilized.

Consequently, the exploration of TP technology in recent years has primarily concentrated on modulating the interface adhesion and stamp materials to amplify the magnitude difference between these forces. This modulation is necessary to meet the requirements of competing fractures for successful pick-up and print processes. This review systematically summarizes these recently developed techniques based on different means of improvement, including (1) the donor/device interface, (2) the device/stamp interface and (3) the stamp choice. The advantages, disadvantages, applicable materials and potential applications of each technique will also be succinctly discussed and sumarized in table 1. Our aim is to distill the latest advancements in TP and provideed valuable guidance for the future design of state-of-art TP technologies with enhanced quality from these three perspectives. Moreover, we aspire to stimulate innovative solutions to address critical challenges in this field, such as mass production capabilities and precision alignment.

Table 1. Comparison of typical TP methods.

Methods	Advantages	Disadvantages	Applicable inks	Applicable substrates	Resolution	Throughput	Cost	Typical applications	References
				Donor/device interface					
Water	Operated at room temperature, easy operation, detects free	Not applicable to water-sensitive materials and substrates, water residue	Thin film nanoelectronics, 2D materials, organic materials	Various flexible substrates	Nanoscale	Wafer-scale	Low	CMOS systems, sensors, and optoelectronics	[58, 70]
SAMs	Easy operation, no influence on devices	High requirements for donor substrates and environments	Metals, oxides, semiconductors, polymers	Various flexible substrates	Sub-100 nm	Wafer-scale	Low	Displayers, micro-LEDs, supercapacitors	[60, 71]
Graphene	High yield, enhance the vdW junctions	Low yields because of defects in graphene	Metals	Graphene, MoS_2, SiO_2	Sub-100 nm	Wafer-scale	High	Semiconductor transistors, FETs	[59, 72]
Sacrificial layer	Near-zero adhesion force	Not applicable to solvent-sensitive materials and substrates, complex processes, residue of solvents	All kinds of materials with suitable sacrificial layers and donor substrates	Various flexible substrates	50 nm	Wafer-scale	Low	Electrodes, micro-LEDs	[61]
				Device/stamp interface					
Contact area	Large adhesion switchability, able to realize 3D alignment	Hard to fabricate the stamp, not applicable to nano- or micro-scale materials and soft substrates	Films	Hard surface	50 μm	Low	Medium	Transistors	[62, 63]

(Continued.)

Table 1. (Continued.)

Magnetism	Continues adjustments of adhesion force	Inhomogeneous deformation of stamps, expensive devices, low accuracy	Relatively rigid materials, films	Various flexible substrates	Microscale	Low	High	Micro-LED display	[73, 74]
Laser	Large adhesion switchability, non-contact model	Expensive machines, possible heat damage to the inks	Materials have heat stability	Various flexible substrates including 3D surface	High	Medium	High	Electronic devices, optical devices	[75, 76]
				Special stamp					
Thermal	Easy operation, low cost	Potential surface residues, thermal damage to the devices	Materials have heat stability	Various flexible substrates	20 nm	8-inch	Low	Photodetectors, flexible strain sensors	[66, 77]
Ice	Ultra-clean transfer	Harsh low-temperature environment, unstable transfer yield	2D materials film	Both hard and flexible substrates	Nanoscale	Medium	Low	2D photoelectronic devices	[67]
Reflowable	Easy operation, high precision, easy alignment	Solvents and heat utilized may limit the kinds of inks	Nanoscale metals (dots, lines, films)	Nanoscale 3D surface, bio-surface-like cells	Nanoscale	Low	Low	Not mentioned	[68]
3D curved	Applicable for 3D curved surface	Only applicable to the special receiver substrate	Si arrays, metals	3D curved surface	Microscale	Medium	Low	Antennas, micro-LEDs, optic devices	[69, 78]

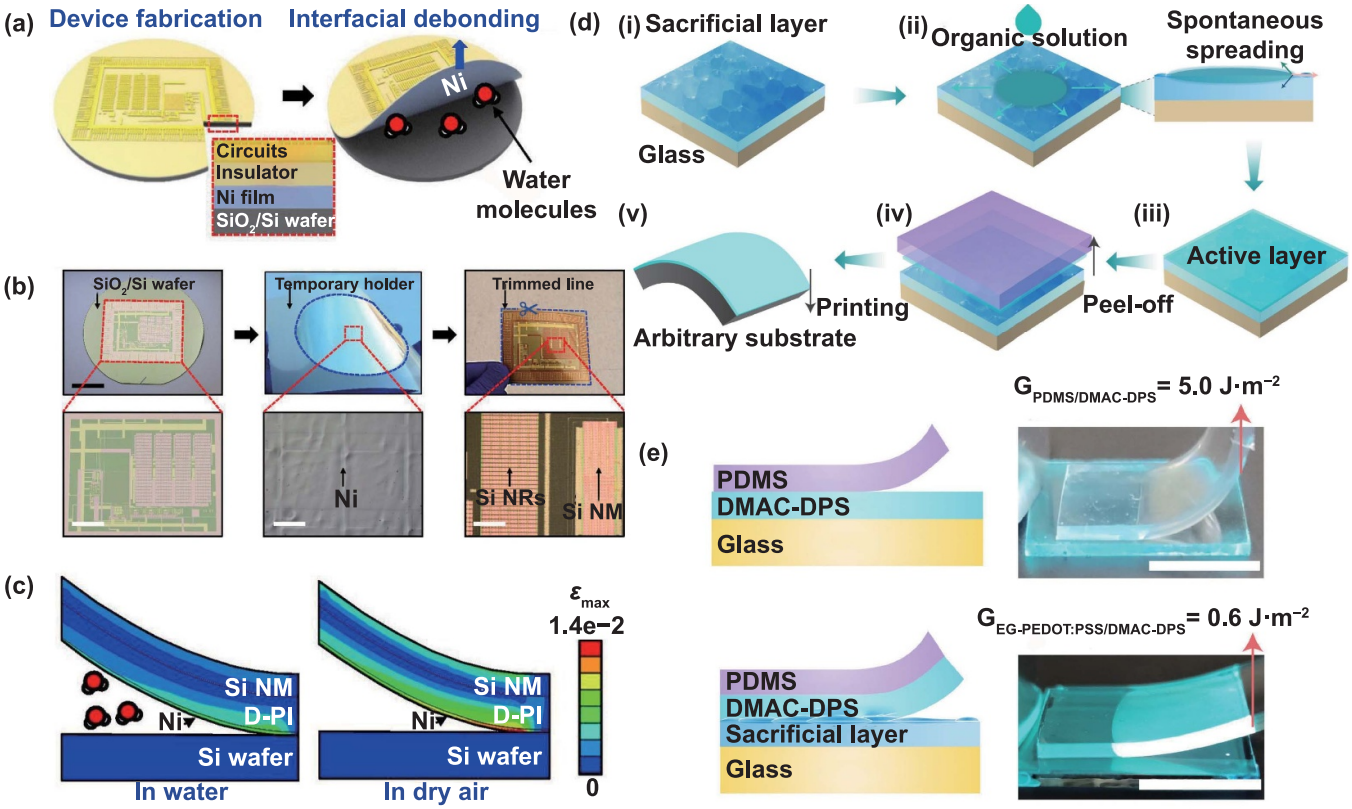

Figure 2. Schematic illustration of water-assisted TP. (a) Utilizing the oxidation of the Ni layer. The devices were fabricated on a SiO_2/Si wafer with a thin layer of Ni film and deboned in water. (b) Optical images of the wafer-scale fabrication of thin-film electronics. Scale bars: (Top) 2.5 cm, (Bottom) 1 cm, 400 and 300 μm, respectively. (c) Finite-element analysis (FEA) results of the theoretical maximum strain distributions on the interface during the peeling process with (left) and without (right) water assistance. Reproduced with permission from [58], © 2018. Published under the PNAS license. (d) Utilizing the hydrophobicity property. (i) Spin-coat of water-rich sacrificial layer. (ii) spontaneous spreading of the organic solution. (iii) spin coat to form a homogenous active layer. (iv) peel off the organic active layer with PDMS. (v) Print of the organic active layer on the arbitrary substrate. (e) Left: illustration of the peeling process without (up) and with (down) hydrophobic layer. Right: the corresponding digital images. Scale bar: 10 mm. Reprinted from [86], © 2023 The Author(s).

2. Methods to modulate transfer printing

The traditional TP method, based on the kinetic control of the elastomeric stamp, provides a straightforward tool for fabricating various hybrid device systems on flexible substrates. However, the limited tunablility of adhesion constrains the selection of applicable materials and substrates, particularly when the receiver is also an elastomer [34]. As previously mentioned, the demand for competing fracture necessitates a specific relationship between the adhesion force of different interfaces. Consequently, numerous efforts have been dedicated to adjusting the adhesion force between these interfaces to satisfy the competitive relationship between the energy release rate between the interfaces. Additionally, the exploration of new types of stamps offers alternative avenues for improving TP.

2.1. Adjustments at the donor/device interface

Aiming at the first step of TP, the pick-up process, the most straightforward approach is to reduce the adhesion force between the donor/device interface, F_1. Various materials or external treatments can be utilized to achieve this, including chemical treatment [79], electrostatic forces [80, 81], electromagnetic forces [73] and vacuum adsorption [82]. In this section, four typical assist methods, including water, self-assembly monolayers (SAMs), graphene and sacrificial layer, will be introduced in detail, supplemented by recent literature.

2.1.1. Water-assisted methods. Water can react with certain metals or metal oxides, e.g. nickel (Ni), aluminium (Al), etc, to form soluble hydroxide [83, 84]. This reaction can be harnessed as a releasing mechanism in TP. For instance, as depicted in figure 2(a), a thin layer of Ni may be predeposited on the conventional SiO_2/Si substrate, with devices subsequently fabricated on this Ni layer. The Ni is anticipated to react and form either nickel silicate or nickel oxide at the interface. Upon exposure to water, the nickel oxide layer is rapidly penetrated, leading to the formation of a hydrophilic layer of nickel hydroxide [85]. This process facilitates the easy detachment of the Ni layer, along with the devices, from the SiO_2/Si substrate using a PDMS stamp. The peeling strength exhibited a significant decrease from approximately 79 $J{\cdot}m^{-2}$ in dry air to about 14 $J{\cdot}m^{-2}$ when facilitated by water [58]. The wafer-scale transfer of thin film electronics

could be realized through the application of a heat-release tape, as shown in the optical images in figure 2(b). Accompanied by the finite elements analysis (FEA) results shown in figure 2(c), the strain distribution in water during the interfacial debonding process is also about 40% smaller than that in ambient air. The decrease in interfacial debonding strength and strain enables the transfer of flexible and thin-film-based devices to any chosen receiver substrates.

Another property related to water that can be exploited is the hydrophobicity of materials. For instance, small organic semiconductor molecules, which are a promising candidate for flexible organic optoelectronic devices, often pose a challenge during transfer from a glass substrate due to uneven film formation and high surface adhesion [86]. However, the adhesion force can be significantly mitigated by utilizing a water-rich releasing layer. This layer comprises a poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT: PSS) layer, doped with high-boiling-point ethylene glycol. The high surface energy of this water solution film enables the spontaneous spreading of hydrophobic small molecule solutions [87]. Consequently, a uniform film of the small molecule layer can be formed and easily detached from the water-rich layer, due to the reduction in van der Waals force and interfacial friction resulting from the hydrophobic characteristic of these organic films (figure 2(d)). The peeling strength between the water-rich releasing layer and the small molecule layer is a mere $\sim$0.6 J·m^{-2}, significantly less than that between the thin film and the PDMS stamp (figure 2(e)). All-organic photodetectors fabricated by this method can be transferred to any flexible substrates, exhibiting exceptional performance features such as repairability and recyclability [88].

Water-assist TP, which can be easily implemented at room temperature and is compatible with traditional substrates, provides significant benefits in preserving the structure and performance of devices. Water-assist TP facilitates a wafer-scale transfer process for a wide range of materials, including metals, nanowires, and nanoscale thin films, among others, at a low cost [70, 83, 89]. Water-assist TP has proven to be highly efficient in the fabrication of various thin film-based electronics such as nanowire circuits and photodetectors. However, the requirements of a specific assisting layer to achieve the releasing function may complicate the fabrication process. Moreover, it may not be applicable to some water-sensitive materials and substrates.

2.1.2. Self-assembly monolayers-modified methods. SAMs are formed through physical or chemical adhesion between organic molecules and the substrate surface [90, 91]. In certain instances, one terminal of the molecule can form strong bonds with the substrate (e.g. thiols, silanes, phosphonates), while the other terminal with different groups serves as an adjustable functional group [92–94].

By capitalizing on their surface modification capabilities, these SAMs can modulate the adhesion force at the substrate/device interface. Gold (Au), a commonly used metal in semiconductors, can form SAMs through thiol terminals [92]. Utilizing this property, Zheng's group introduced a novel patterning method known as electrochemical replication and transfer (ERT) [71, 95]. This method, schematically represented in figure 3(a), commences with a Cr/Au patterned template fabricated using traditional lithography methods. A SAM of 1H,1H,2H,2H-perfluorodecanethiol (PFDT) is then grown on the Au surface, transforming it from hydrophilic to hydrophobic (figure 3(b)). This modified template is employed in the subsequent electroplating process, resulting in the growth of a target material layer on the Au pattern. The electrodeposited layer is then separated from the Au template and transferred to a target flexible substrate using a binder layer. The successful separation of the metal pattern from the Au template relies heavily on the PFDT SAM modification. As shown in figures 3(c) and (d), the Cu pattern and Au template could remain intact only with the modification of PFDT. This low-surface-energy SAM layer reduces the adhesion force and protects the Au pattern, enabling the reuse of the Au template (figures 3(e) and (f)). Various materials, including metals, oxides, semiconductors, and polymers, are all compatible with the ERT process. It has proven effective in fabricating various high-resolution flexible and stretchable electronic devices reaching sub-100 nm like OLEDs and supercapacitors on different substrates.

In addition to modifying Au metal patterns, silane SAMs can also be employed to modify pure Si substrates. For example, octadecyl trichlorosilane SAMs can form a dense layer on the Si substrate via a robust covalent bond [96]. This surface modification reduces the surface energy from 1140–21 mJ·m^{-2} [97]. The modified Si substrate is particularly benificial for the transfer of quantum dots (QDs), a material that exhibits optimal performance when prepared via spin coating. Traditional patterning methods for fabricating QDs on flexible substrates, such as contact printing and mist deposition, often enconter difficulties in achieving a uniform film and superior performance [98–100]. However, with the aid of SAMs, TP can effectively pattern high-quality QD films without causing damage (figure 4) [60]. Large-area, patternable, flexible, full-colour QD displays can be easily fabricated with enhanced performance.

The surface modification of SAMs occurs at the molecular level, minimally affecting the TP process and device fabrication. Moreover, the modification process is straightforward and devoid of side effects. However, potential hazards exist in the form of defects in the SAM layer. Externally, the self-assembly process requires a high degree of substrate cleanliness, as impurities and environmental changes can influence molecule formation. Intrinsically, etch pits (vacancies) may form due to the thermodynamics of formation [90]. These defects could potentially impact the yield of the TP process.

2.1.3. Graphene-assisted methods. Graphene, a well-known 2D material, possesses a uniform surface characterized by weak van der Waals forces [101]. In conventional metal deposition processes on semiconductor surfaces (Si/SiO_2), metals can form strong chemical bonds with the substrates. This is particularly true for high-adhesion metals such as

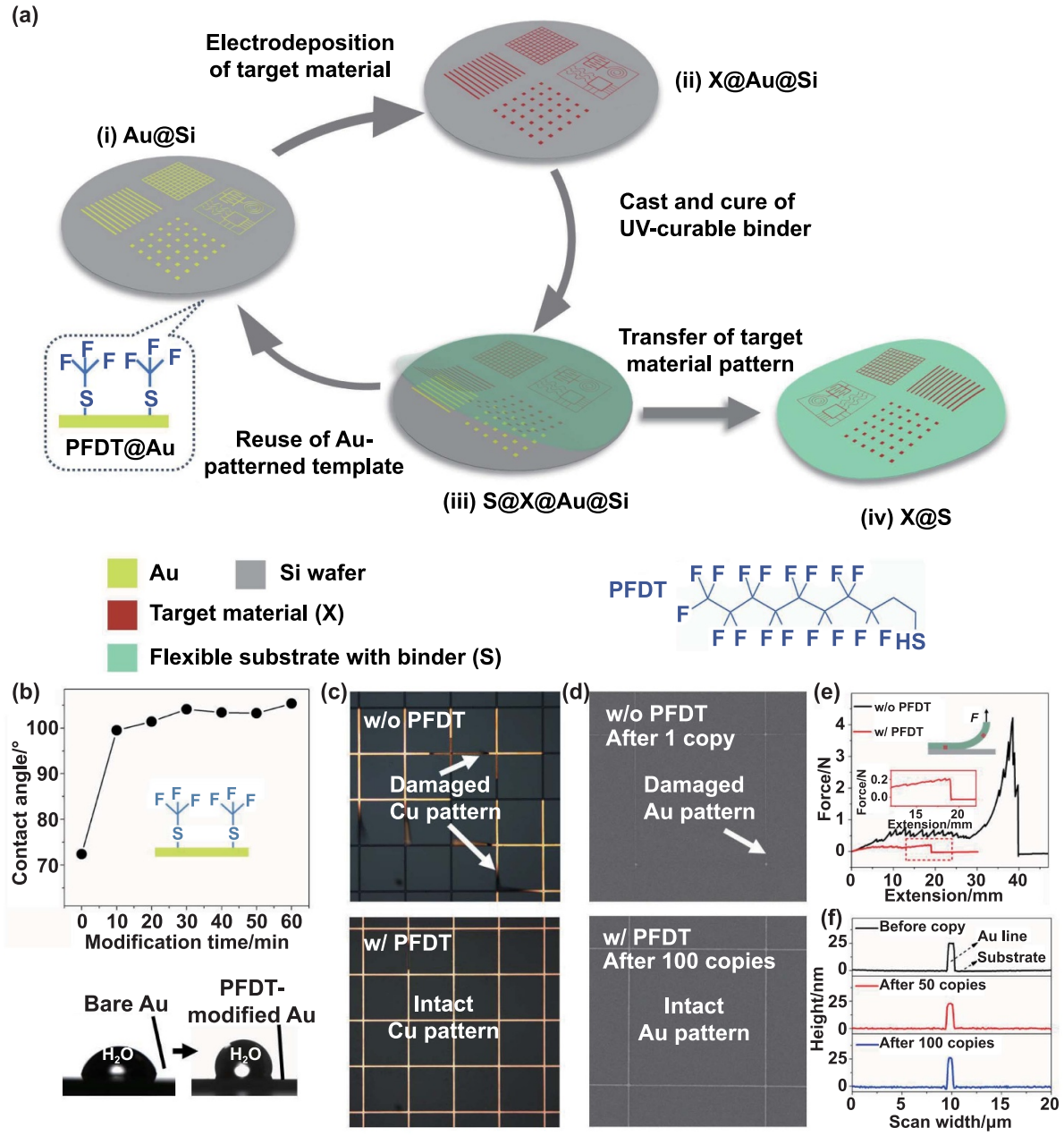

Figure 3. Schematic illustration of Electrochemical Replication and Transfer (ERT). (a) Fabrication process of ERT. (i) Modification of Au patterned template with PFDT solution. (ii) Electrodeposition of target materials. (iii) Cast and cure of UV curable binder. (iv) Peel off the target materials pattern and transfer to the target substrate while the Au template could be reusable. (b) The change of contact angle as the decreasing of PFDT modification time. (c) Optical images of the Cu pattern peeled off from the Au template without (up) and with PFDT modification. (d) Optical images of the Au template after 1 fabrication process without PFDT modification (up) and after 100 fabrication process with PFDT modification (down). (e) The peeling force with and without PFDT modification. (f) The surface profiler results of the Au template after 100 times of fabrication. [71] John Wiley & Sons. © 2023 Wiley-VCH GmbH.

platinum (Pt), titanium (Ti), and nickel (Ni), which play a crucial role in forming metal-semiconductor junctions for electronic and optoelectronic devices [102]. However, for TP processes, such strong adhesion force between the donor substrate and metals may result it damage [103].

A potential solution is the introduction of a 2D graphene layer on the surface of the donor substrate. The graphene film can be grown on a Ge substrate through chemical vapour deposition [104], and the patterned metal film can then be deposited onto this Graphene/Ge substrate. Due to the absence of dangling bonds on the graphene surface, these metals can be easily peeled off by a binder layer (e.g. PVA). The yield can reach nearly 100%, even for high-adhesion metals like Pt (figure 5(a)). Assisted by Raman spectra results, it can be confirmed that the metal could be easily detached from the graphene surface without damaging the graphene layer

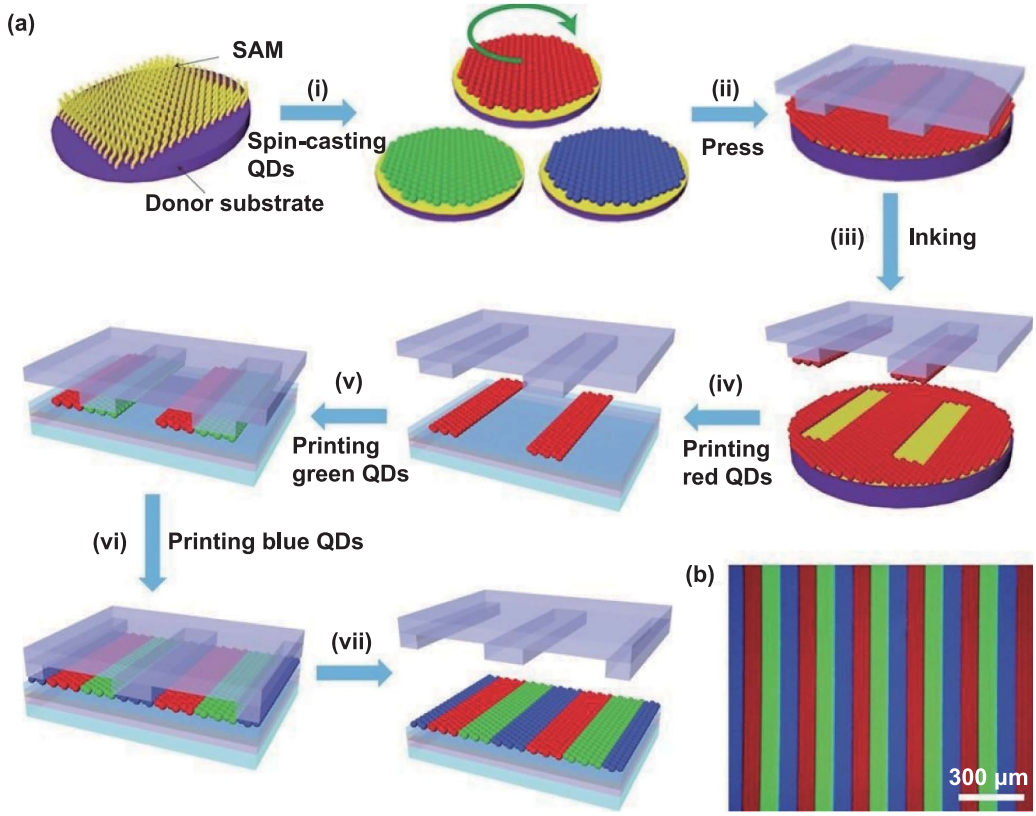

Figure 4. Fabrication of QDs with SAM assist TP. (a) Schematic illustration of the method. (i) Spin-casting of QDs on a SAM-modified donor substrate. (ii) Put the patterned elastomer stamp on the QDs with pressure. (iii) Peel off the QDs. (iv)–(vi) Printing of QDs with different colours successively under alignment. (vii) Remove the elastomer stamp. (b) The fluorescence image of the RGB QDs on the glass substrate. Reproduced from [60], with permission from Springer Nature.

(figure 5(b)). This graphene-assisted TP method can efficiently generate wafer-scale semiconductor transistor arrays with homogeneous electrical performance [59].

Furthermore, the graphene layer can also be transferred in conjunction with the metal layers. As depicted in figure 5(c), the graphene layer is first transferred to a Si substrate, which subsequently acts as the donor substrate in the ensuing TP process. After the patterning and metal deposition process on this graphene/Si substrate, the residual portions of graphene unprotected by metal are etched by oxygen or argon plasma. Subsequently, the patterned metal/graphene devices can be effortlessly peeled off by a poly(methyl methacrylate) (PMMA) layer. The graphene layer, possessing an extremely low van der Waals force at the interface between Si and graphene, can also eliminate the gap status induced by 3D metals, which is beneficial for the application of field-effect transistors [72].

The distinctive properties of 2D materials, particularly graphene, contribute significantly to semiconductor devices. Its unique surface properties provide it with considerable potential in the TP process. However, the primary challenge resides in the uniform fabrication of the graphene layer without defects. The associated costs and specific synthesis conditions may also restrict its application.

2.1.4. Sacrificial layer-assisted methods. In numerous lithography techniques, the application of a sacrificial layer between the substrate and the devices is a prevalent method to separate the devices or unwanted parts. This layer, easily removable or dissolvable in certain solvents, also functions as an effective tool in the pick-up process for TP.

The sacrificial layer can separate the interface through a direct dissolution process. For example, water-soluble dextran can initially be spin-coated onto the Si/SiO_2 wafer surface, acting as the modified substrate for the subsequent photolithography process. After the electrospinning of a fiber mat on the substrate, the patterned metal can be successfully transferred to the flexible fiber mat by dissolving the dextran layer in water, enabling the metal pattern to be transferred to the target flexible substrate without damage [105]. Other materials like poly(acrylic acid) [106] and polyvinyl alcohol [107] can also be used as the sacrificial layer due to their water solubility. This sacrificial layer-assisted method is compatible with traditional lithography methods and allows the target substrate to directly receive the devices from the donor substrate, bypassing the conventional 'print' process, due to the near-zero adhesion force between the donor substrate and the devices after the sacrificial layer is dissolved.

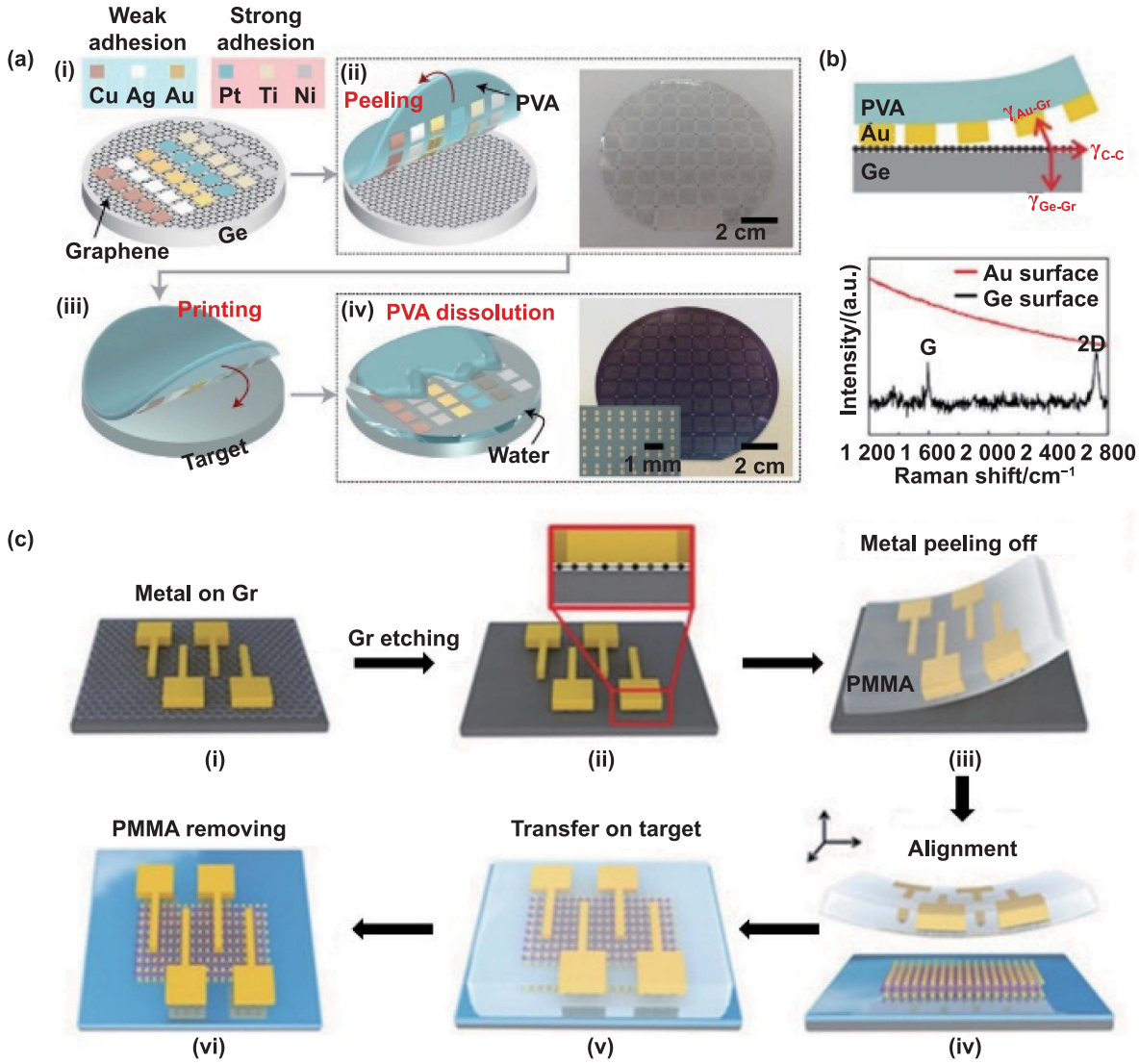

Figure 5. Schematic illustration of graphene-assisted TP. (a) Fabrication process using graphene as the releasing layer. (i) Deposition of target metals onto the Ge substrate with the graphene layer. (ii) Cast of PVA binder and peel off the target metals. (iii) Print the target metals onto the target substrate. (iv) Dissolve the PVA layer with water. (b) Schematic illustration and Raman spectra of the surface after the peel-off process of Au pattern from the substrate with graphene. Reproduced from [59], with permission from Springer Nature. (c) Fabrication process using graphene in conjunction with metal layer. (i) Deposition of metal pattern on the substrate with graphene layer. (ii) Etch the extra graphene without a metal pattern. (iii) Peel off the metal pattern with graphene by PMMA. (iv) Alignment of the metal pattern. (v) Print the metal pattern onto the target substrate. (vi) Dissolve the PMMA layer. [72] John Wiley & Sons. © 2023 Wiley-VCH GmbH.

Another strategy for applying the sacrificial layer capitalizes on the low adhesion force between the sacrificial layer and the donor substrate. As previously mentioned, some metals, such as Cu film [108], exhibit low surface energy, making them easy to peel off from donor substrates. This is demonstrated in figure 6(a), where a thin layer of Cu is pre-deposited on the PMMA substrate. The ink material is then fabricated on the Cu film and peeled off by the stamp. The extremely low energy release rate of the Cu/donor interface aids in increasing the resolution limits from micrometres to nanometers even under a low peel-off speed, i.e. 5 mm·s^{-1}. Additionally, the Cu sacrificial layer serves as an interlayer, and by optimizing the layer thickness, the mechanical neutral plane of the entire device/stamp layer can be adjusted, eliminating fracture strain on the device layer (figures 6(b) and (c)) [109]. In the final step, the Cu layer is chemically etched away, for example, by a $FeCl_3$ solution. The protection provided by the Cu sacrificial layer allows the transferred pattern to achieve a resolution of sub-50 nm on various flexible substrates [61].

The prevalent application of sacrificial layers in TP underscores its significance. The selection of an appropriate sacrificial layer is vital, as the solvent used should neither react with nor dissolve the devices and receiver substrate materials. However, the additional step of dissolving the sacrificial layer may introduce complexity and potential unpredictability to the process, potentially causing damage to the devices.

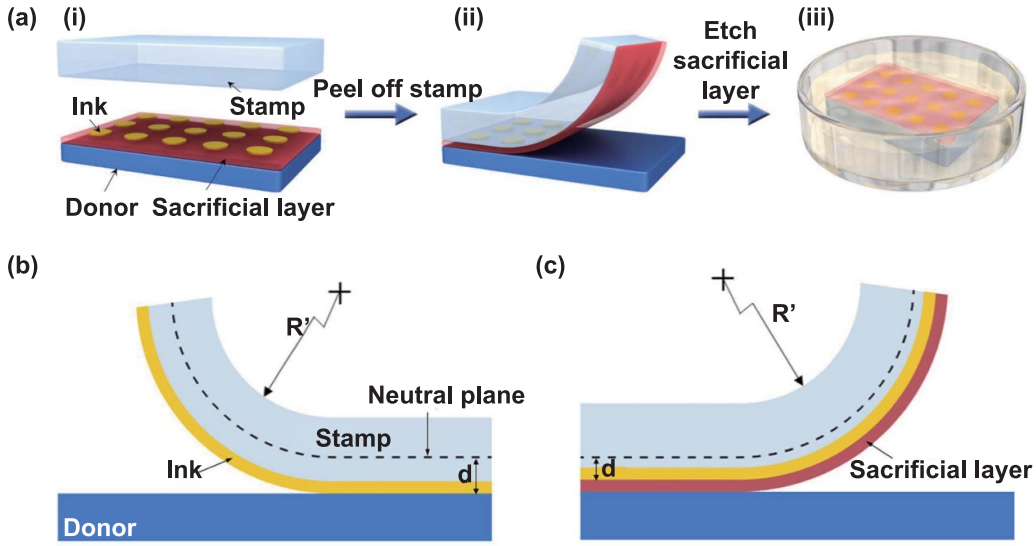

Figure 6. Cu sacrificial layer in TP. (a) Fabrication process. (i) Press the elastomer stamp on the donor substrate with ink materials. Cu serves as the sacrificial layer. (ii) Peel off the stamp with the ink and Cu layer. (iii) Dissolve the Cu sacrificial layer. (b) Diagram and the neutral plane of the peeling process without sacrificial layer. (c) With sacrificial layer. Reproduced from [61]. CC BY 4.0.

2.2. Adjustments at the device/stamp interface

The device/stamp interface is another area susceptible to device breakage. Implementing modifications on this interface to reduce the adhesion force could facilitate the complete transfer of devices onto the receiver substrate without damage. In this section, three typical manipulation methods including contact area, magnetic and laser will be introduced. Other external forces such as electrical and thermal assist methods could also be highly efficient while will not be discussed in detail here [79, 80].

2.2.1. Control of contact area. For the traditional elastomer stamp, the adhesion force between the interfaces is directly proportional to the contact area. Building on this principle, Rogers' group proposed a structured design for the elastomer stamp, drawing inspireation from small animals such as geckos [62]. This design features a stamp surface with pyramid-like microstructures. As demonstrated in figure 7(a), these pyramid tips collapse under high load during the pick-up process, thereby creating a contact area nearly equivalent to that of a flat stamp and ensuring successful peeling off of the devices. Following the pick-up process and load release, the stamp's resilience allows the pyramid tips to regain their original shape. This significantly reduces the contact area between the stamp and the device, as only the tip of pyramid spire comes into contact with the devices. In the subsequent printing process, the stamp bearing the devices contacts the receiver substrate under low pressure. The devices remain on the substrate after the stamp is gently removed. The inserted SEM images depicted the whole deformation process, and the required force was plotted in figures 7(b) and (c). Compared with a flat surface, the microtip structure provided a difference of over three orders of magnitude in the adhesion forces between the interfaces before and after the collapse of the stamp, resulting in a high-yield transfer process (figures 7(d) and (e)). Moreover, this method can easily achieve multilayer stacked patterns on a variety of substrates.

Beyond leveraging the elastomeric properties of stamps, some shape memory materials are also suitable for stamp functions and adjusting the contact area [110, 111]. For instance, decyl amine (NGDE), a thermoset shape memory polymer, exhibits strong adhesion when rigid at a temperature below its glass transition temperature (T_g) [112]. When the temperature rises to the T_g, the polymer becomes elastomeric and recovers its original shape. By creating similar surface microstructures, NGDE can serve as a stamp for TP by adjusting the contact area [63]. Shape memory materials outperform conventional elastomeric materials because shape control can be achieved through heat change rather than time sensitivity.

However, contact area control TP methods may not be suitable for transferring larger area devices. These large devices are susceptible to cracking due to the non-uniform distribution of microstructure surfaces. Stress concentration can easily occur at the microstructure's tip, leading to unpredictable damage to the devices. Besides, the use of micro-tips additionally requires precise contact control to avoid failure caused by excess deformation.

2.2.2. Magnetism-assisted methods. Magnetic force can be employed to modify the adhesion between the device and the stamp interface. This is demonstrated by the special structure design of the stamp [74]. The stamp, a columnar object, features a magnetic-responsive thin film on its top. An external magnetic field can remotely manipulate this film, causing it to either bulge or sag. This change in the stamp cavity's volume alters the pressure, which in turn adjusts the adhesion force between the stamp's bottom surface and the devices. This regulatory mechanism enables a continuous modulation process of the interfacial adhesion force.

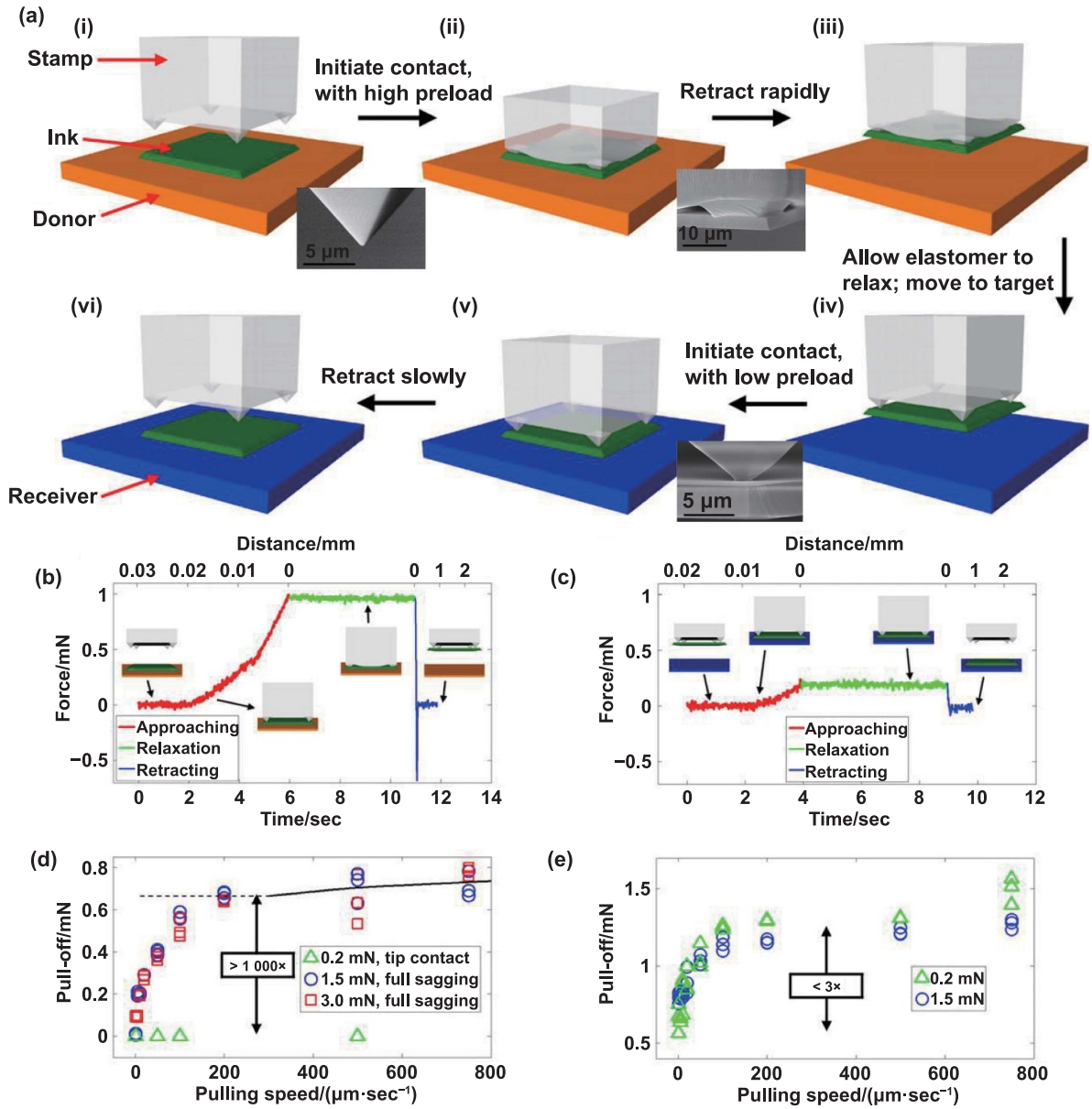

Figure 7. TP using an elastomer stamp with micro tips on the surface. (a) Fabrication process. (i) Place the stamp on the donor substrate with ink. (ii) Press the stamp with a high load. (iii) Retract the stamp quickly. (iv) Place the stamp with ink on the receiver substrate after the relaxation of the stamp. (v) Press the stamp with a low load. (vi) Retract the stamp slowly. (b) Force–Time–Distance relationship during the pick-up process. (c) Force–Time–Distance relationship during the print process. (d) The pull-off force of a microtip surface from the silicon. (e) The pull-off force of a flat surface from the silicon. Reproduced from [62]. © 2010. Published under the PNAS license.

A variety of effective stamp designs can be fabricated to accommodate different substrates and materials, as demonstrated through theoretical analysis and numerical calculations of stamp structure [65]. Recently, Cheng's group introduced an innovative approach to TP by incorporating NdFeB (Neodymium Iron Boron) magnetic particles into a silicone layer to create a magnetic stamp film [73]. Through establishing a theoretical model by FEA, the deformation of the magnetic film could be analyzed and predicted. The deformation process could be well controlled by adjusting the mass fraction of magnetic particles, film thickness and magnetic field strength, therefore controlling the adhesion force during the TP.

The exploration of magnetism-assisted TP has predominantly revolved around theoretical optimization. A range of theoretical models, encompassing both two-dimensional and three-dimensional mechanical frameworks, have been developed to guide the adaptation process for a diverse array of materials [113, 114]. The specialized design of the magnetic film has also broadened the spectrum of compatible substrates, extending to static, flexible, and porous structures.

Despite the efficiency and control offered by this method, there are challenges to be addressed. The potential for inhomogeneous deformation in the magnetic film could hinder its scalability and compromise the accuracy of alignment [115]. Moreover, the cost implications of this technique and the need

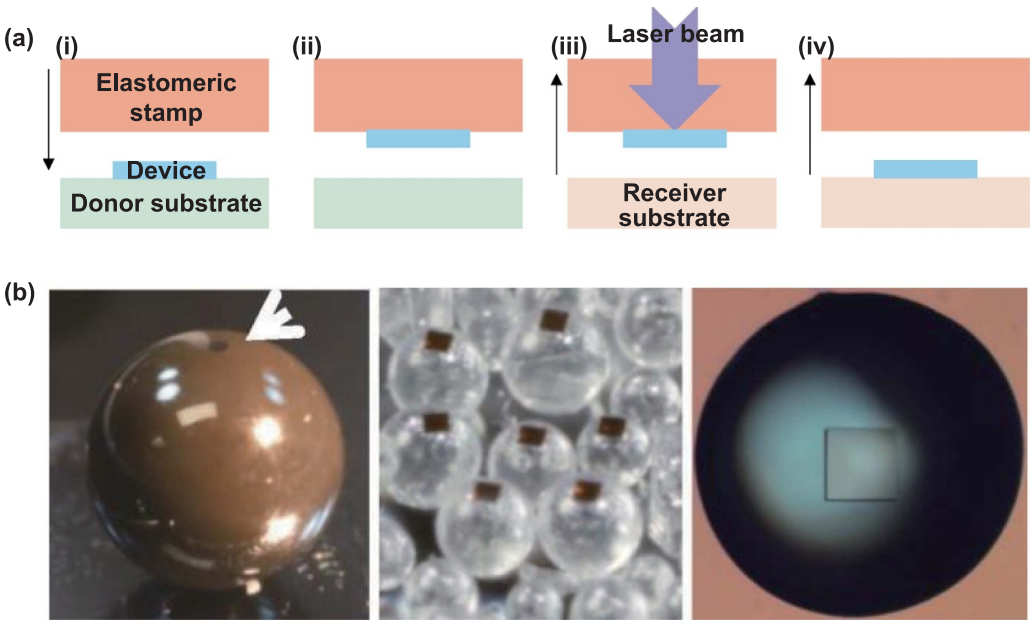

Figure 8. Laser assist TP. (a) Schematic illustration of the process. (i) Place the stamp on the donor substrate with devices. (ii) Pick up the device quickly. (iii) Applying a laser beam to release the devices. (iv) Remove the stamp. (b) Printing on a 3D curved surface. (a) and (b) Reprinted from [76], Copyright © 2012 The Society of Manufacturing Engineers. Published by Elsevier Ltd. All rights reserved.

for specific environmental conditions may limit its practical application [115].

2.2.3. Laser-assisted methods. Laser is another potent external source that can be utilized for TP. By modulating parameters such as wavelength, intensity, and reaction time, different physical or chemical reactions can occur at the interface between the stamp and devices. These reactions may include deformation, melting, decomposition, and phase/-structural transformation [35, 116–118]. Laser irradiation can induce a broad range of adjustments at the device/stamp interface, making it suitable for a variety of materials.

In the case of classic elastomer stamps like PDMS, their transparency allows a pulsed laser beam to pass through the stamp to the device/stamp interface. Instead of causing physical or chemical damage, the pulsed laser increases the device's temperature. This heat is transferred to the PDMS stamp, raising its temperature as well. However, the differing thermal responses of the stamp and devices lead to distinct deformations, which subsequently trigger a gradual, automatic detachment of the devices from their edge. This process is schematically illustrated in figure 8(a) [75] and has been confirmed via high-speed camera observations and theoretical calculations [75, 119, 120]. Since laser TP allows powerful manipulation of the adhesion force down to near zero on the device/stamp interface, it enables a non-contact transfer model in addition to the traditional contact and proximity transfer model, which significantly broadens the range of receiver substrates, including complex 3D surfaces (figure 8(b)) [76]. It also boasts high accuracy, quick response, spatial resolution, and area controllability. Furthermore, the non-contact transfer characteristic makes the use of an elastomer stamp unnecessary for some transparent substrates like fused silica, enabling a direct transfer from the donor substrate to the receiver substrate [64].

In this process, the transparent substrate functions as both the donor substrate and the stamp. A dynamic release layer is used between the donor substrate and the devices. The laser's interaction with the dynamic release layer could generate bubbles that provide the force required to release the devices from the reaction layer. This gentle printing process ensures that the devices remain undamaged. Other materials that can react with lasers, such as the thermal decomposition of GaN [121], ablation of Polyimide [122], melting of $Pb[Zr_{0.52}Ti_{0.48}]O_3$ [123], and melting of 1-Si: H [124], can also serve as the releasing layer. The throughput of laser-assisted TP can be significantly increased by integrating parallel laser sources, enabling a high assembly rate of over 100 M units per hour [125].

In addition to leveraging a release layer, lasers can be employed directly on target materials. For instance, the application of a femtosecond laser, capable of delivering high energy in an extremely brief duration, has the potential to induce exfoliation or prompt physical and chemical reactions in a variety of substrates. This technique can be particularly effective for patterning common metals, such as gold (Au) particles, as well as other materials that are sensitive to laser exposure, e.g. $Ge_2Sb_2Te_5$. This direct laser application opens up possibilities for precise patterning without the need for traditional photolithographic processes, potentially streamlining the fabrication of intricate structures on a microscopic scale [126, 127]. It has been proven to be efficient in the fabrication of various optics devices [128].

Laser-assisted TP boasts compatibility with a wide range of materials and substrates that are amenable to conventional

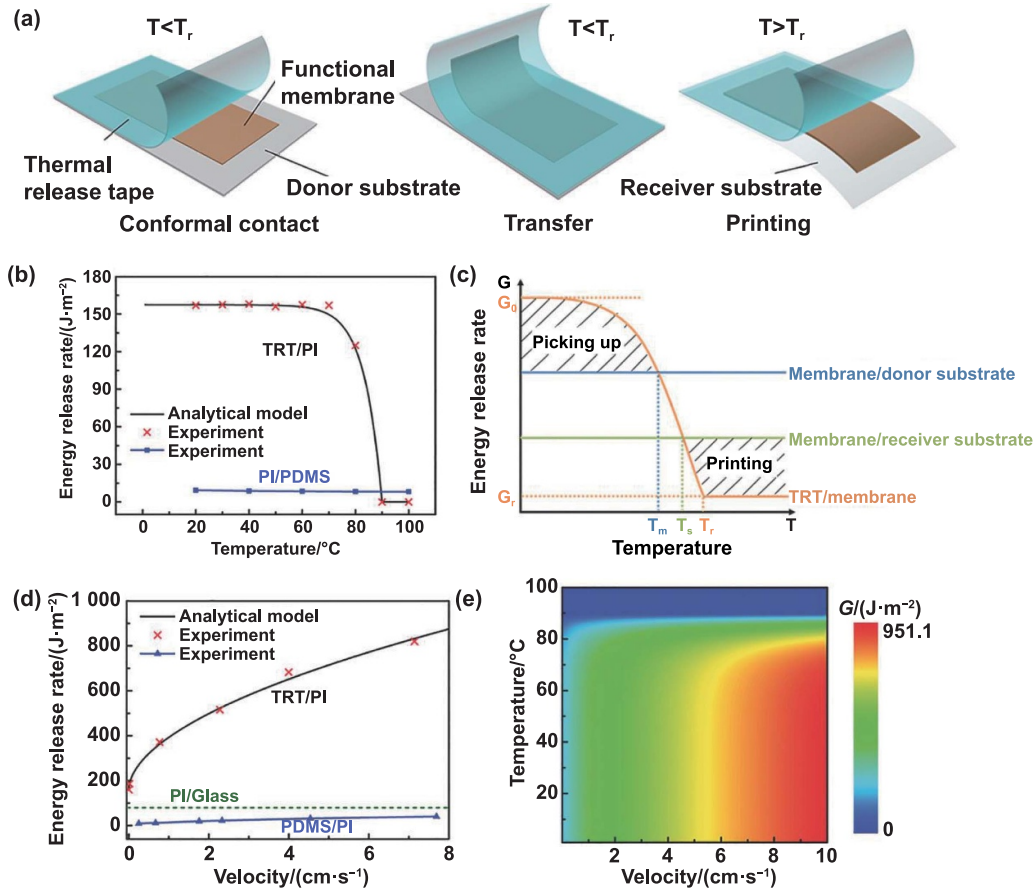

Figure 9. Mechanism of thermal assisted TP. (a) Schematic illustration of the process. (b) Experimental and theoretical calculations of the relationship between temperature and energy release rate. (c) The influence of temperature in the TP process. (d) Experimental and theoretical calculations of the relationship between velocity and energy release rate. (e) The contour map of the relationship between temperature, velocity and energy release rate. (a)–(e) Reproduced from [66]. CC BY 4.0.

elastomeric stamps. Additionally, it offers the advantage of maintaining high resolution and accuracy, making it a versatile and precise option for TP applications. Despite its advantages, it also has a notable limitation: the requirement for expensive equipment to precisely control the intensity and exposure time of the lasing pulse. Additionally, the parameters for each material vary, necessitating extensive adjustments.

2.3. Special stamp design

In addition to various approaches to interface adjustment, classical elastomer stamps have also been supplemented by an extended category of TP stamps to compensate for their limitations in the selection of materials and substrates. The special stamps employ diverse mechanisms to regulate the interfacial adhesion with the devices, thereby enabling high-quality lossless transfers and expanding the range of application scenarios. The TP onto complex 3D structures is also realized through the elaborate design of the stamps. This section will discuss specialized stamp materials such as heat release tape, ice, sugar, balloon, and wrap-like stamps, which are tailored to specific application scenarios rather than the adjustment of interfacial adhesion. It should be noted that the unique characteristics of these stamps may restrict their use with numerous materials and conventional flexible substrates. Additionally, their resolution capabilities and the full spectrum of their potential applications require further exploration and enhancement to fully realize their benefits in the broader context of TP technology.

2.3.1. Thermal-assisted methods. Thermal release tape is a commonly used stamp in TP processes. The adhesion strength of thermal release tape significantly decreases when the temperature rises above the tape's transition temperature [66]. The tape's flexibility facilitates easy operation and a conformal peel-off and print process. By combining kinetic and thermal control, a high ratio of adhesion force between the peel-off and print process can be achieved, enhancing the success rate of transfer (figure 9(a)) [129]. The efficacy of thermal release tape is demonstrated in the experimental results and theoretical calculations shown in figures 9(b)–(e) [66]. The heat plays a crucial role in the changing energy release rate between the interfaces. Upon elevating the temperature beyond 70 °C, there is a marked decline in the energy release rate, plunging from in excess of 150 J·m^{-2} to nearly zero. This dramatic transition, in conjunction with alterations in the peeling speed, synergistically contributes to the efficacy of the

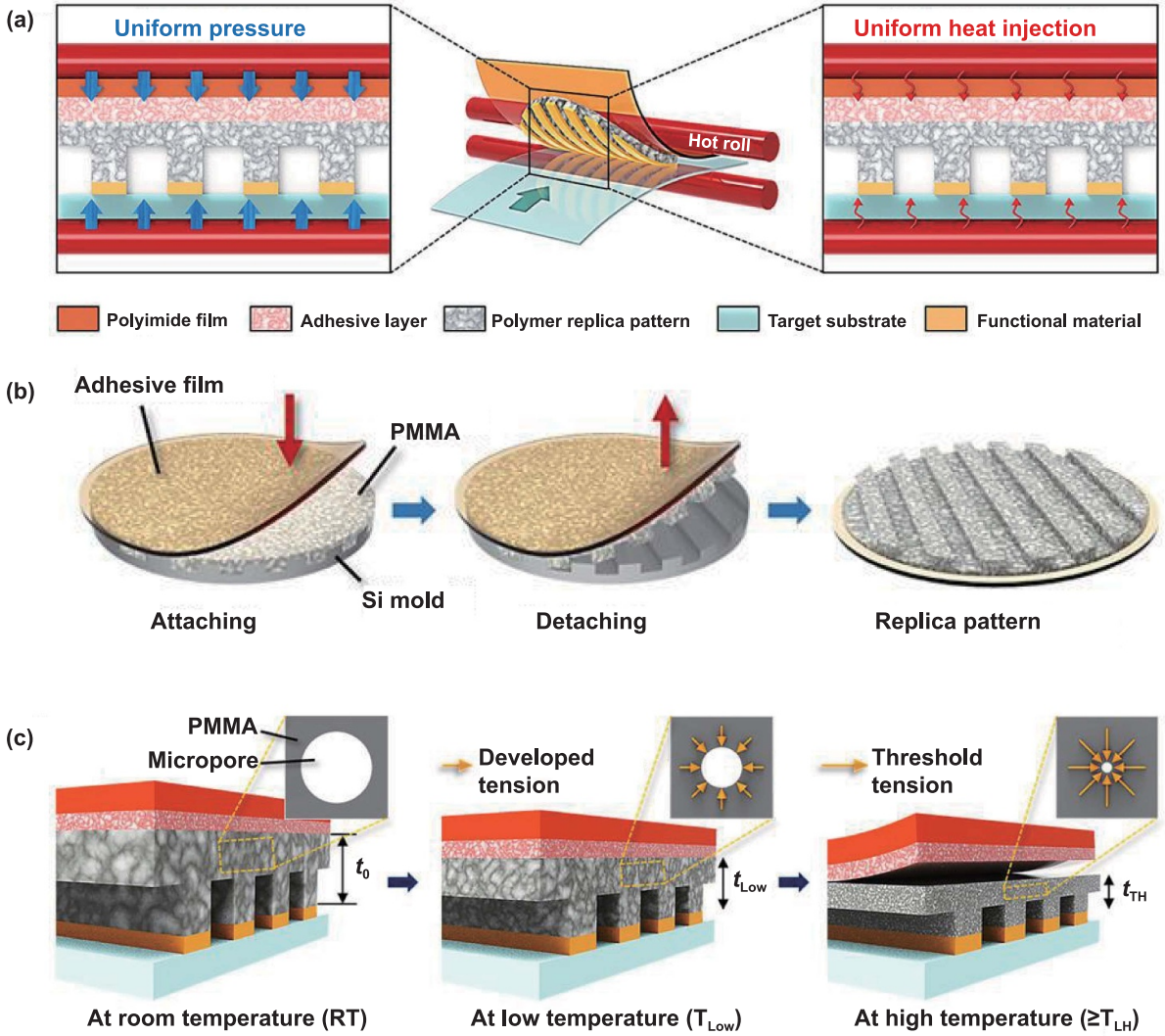

Figure 10. Thermal assist nano TP. (a) Schematic illustration of the roll-to-roll printing process. (b) Fabrication of PMMA mould. (c) Mechanism of the thermal induced TP process. (a)–(c) From [77]. Reprinted with permission from AAAS.

transfer process. Such a temperature-dependent modulation of adhesion properties is instrumental in the precise control of TP, enabling the successful detachment and subsequent placement of materials onto target substrates.

Due to the simplicity and cost-effectiveness of using heat in manufacturing, thermal assist TP can easily be integrated with roll-to-roll fabrication to enable large-area printing techniques [77]. As demonstrated in figure 10(a), a heat-rolling press system was used to ensure uniform heat transfer and applied pressure during the thermal-assisted TP process. A patterned PMMA stamp layer was created by simply spin coating on a pre-fabricated hydrophobic Si mold. After being peeled off by a PI film with an adhesive layer, the patterned PMMA layer could be transferred to the target substrate through heating (figure 10(b)). The porous PMMA layer shrinks upon heating, reducing the adhesion force between the PMMA and the adhesive layer (figure 10(c)). The hot roll plays a crucial role in applying uniform pressure to prevent microcracks and void defects that might arise from manual printing processes. Using the same system and PMMA mold, a variety of metallic and semiconducting materials with high-resolution nano-scale patterns (less than 20 nm) over a large area (up to 8 inches) can be fabricated on flexible substrates like PET.

Other thermal-responsive materials can also be used in thermal assist TP. For instance, Song's group used a shape-conformal thin polymer embedded with commercial thermal expandable microspheres as a stamp to facilitate the TP process [130]. At room temperature, the microspheres are embedded in the polymer layer, maintaining a flat surface that does not affect the adhesion force between the polymer stamp and the target materials. When the stamp is heated to a certain degree (e.g. 90 °C), the microspheres expand, inducing surface hierarchical microstructures and significantly reducing the adhesion force. This process is programmable and scalable and has proven to be an efficient method for fabricating flexible inorganic electronics, such as Si nanomembrane-based photodetectors and flexible strain sensors.

The simplicity and cost-effectiveness of heat-assisted TP have led to its widespread adoption across numerous applications. The incorporation of thermal release tape, in

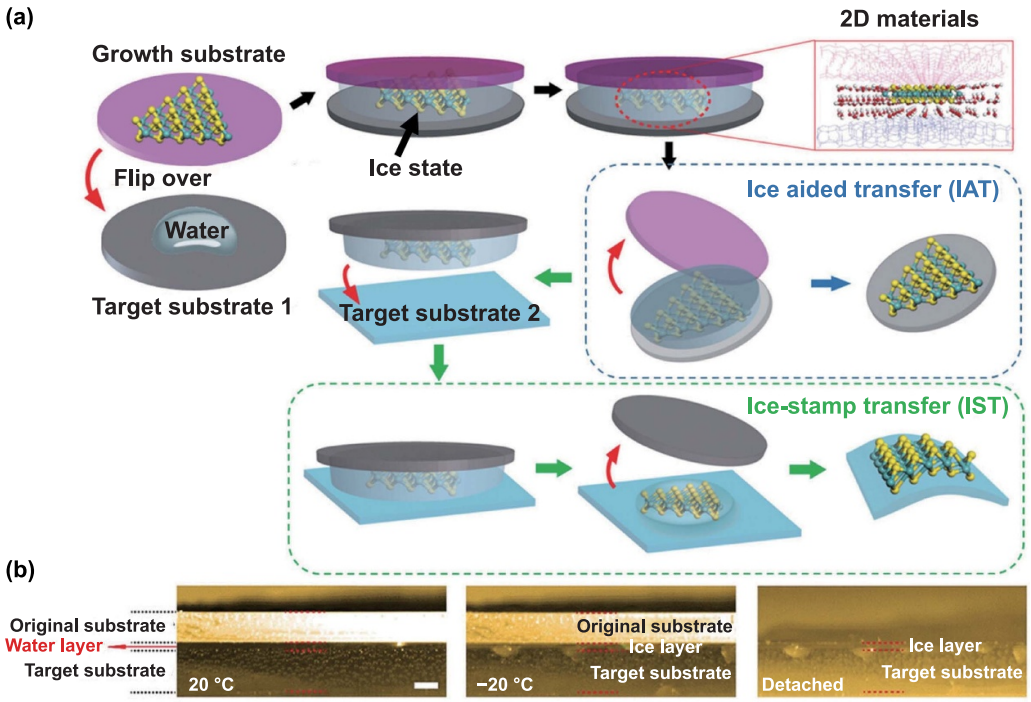

Figure 11. Schematic illustration of ice-assisted TP. (a)The route could be ice-aided transfer (IAT) or ice-stamp transfer (IST) for different substrates. (b) Optical images of the cross-section area during the 'attach', 'freezing', and 'detach' processes. (a) and (b) [67] John Wiley & Sons. © 2023 Wiley-VCH GmbH.

particular, represents a mature commercial product that has streamlined this process. Despite these advantages, the application of heat presents a risk of damage to thermally sensitive materials. Additionally, the process can leave behind residues from adhesives and heat effects on the device surface, potentially compromising device performance. These issues highlight the need for careful consideration of material properties and post-transfer cleaning procedures in heat-assisted TP methodologies.

2.3.2. Ice-assisted methods. Transfer yield is important, so is the quality of the transferred materials, particularly in the context of nanoscale devices such as those involving the transfer of 2D materials. Maintaining cleanliness during the transfer, thereby avoiding contamination, is a significant quality indicator. To this end, the use of ice (water) has been proposed as a highly effective tool for the clean TP of 2D materials [67]. The entire ice-aided transfer process is depicted in figure 11(a). It begins with the addition of an ultra-pure water droplet onto the target substrate's surface, which is then covered with a two-dimensional material. The entire substrate is cooled to freeze the water. Subsequently, the growth substrate is removed, leaving both the two-dimensional material and ice on the target substrate's surface. The process concludes with the application of heat to melt the ice, thereby completing the transfer. The cross-section images during the whole 'attach', 'freezing', and 'detach' processes were listed in figure 11(b).

An ice-assisted secondary transfer process could be implemented using ice as the stamp for target hydrophobic or non-uniform substrates, such as flexible substrates. A key element of this transfer process is controlling the adhesion of ice. Specifically, it is crucial to ensure that the adhesion between the ice and the target substrate surpasses that between the ice and the originating substrate, and that the adhesion between the ice and the 2D material is stronger than the van der Waals interactions between the 2D material and its original substrate.

These adhesive forces can be regulated by manipulating both the temperature of the liquid water and the freezing temperature, making this process adaptable for transferring a wide variety of substrates and two-dimensional materials [131–133]. This adaptability could also be used to clean 2D materials and enhance their long-term stability. The surface of the sample obtained via this method is extremely flat, with no residue due to ice's dissolvability. This method's efficacy is evidenced by the successful transfer of 2D MoS_2 to a flexible PET substrate, which demonstrated excellent photoelectric performance. This outcome can be attributed to the use of ice instead of an organic solvent, which prevents damage to the surface of flexible substrates [134]. A notable challenge with ice-assisted TP lies in the inherently low adhesion and switchable nature of ice, which can adversely affect the transfer yield. Achieving a 100% transfer yield for various 2D materials remains a difficult endeavor due to these limitations. Moreover, when it comes to more conventional materials, such

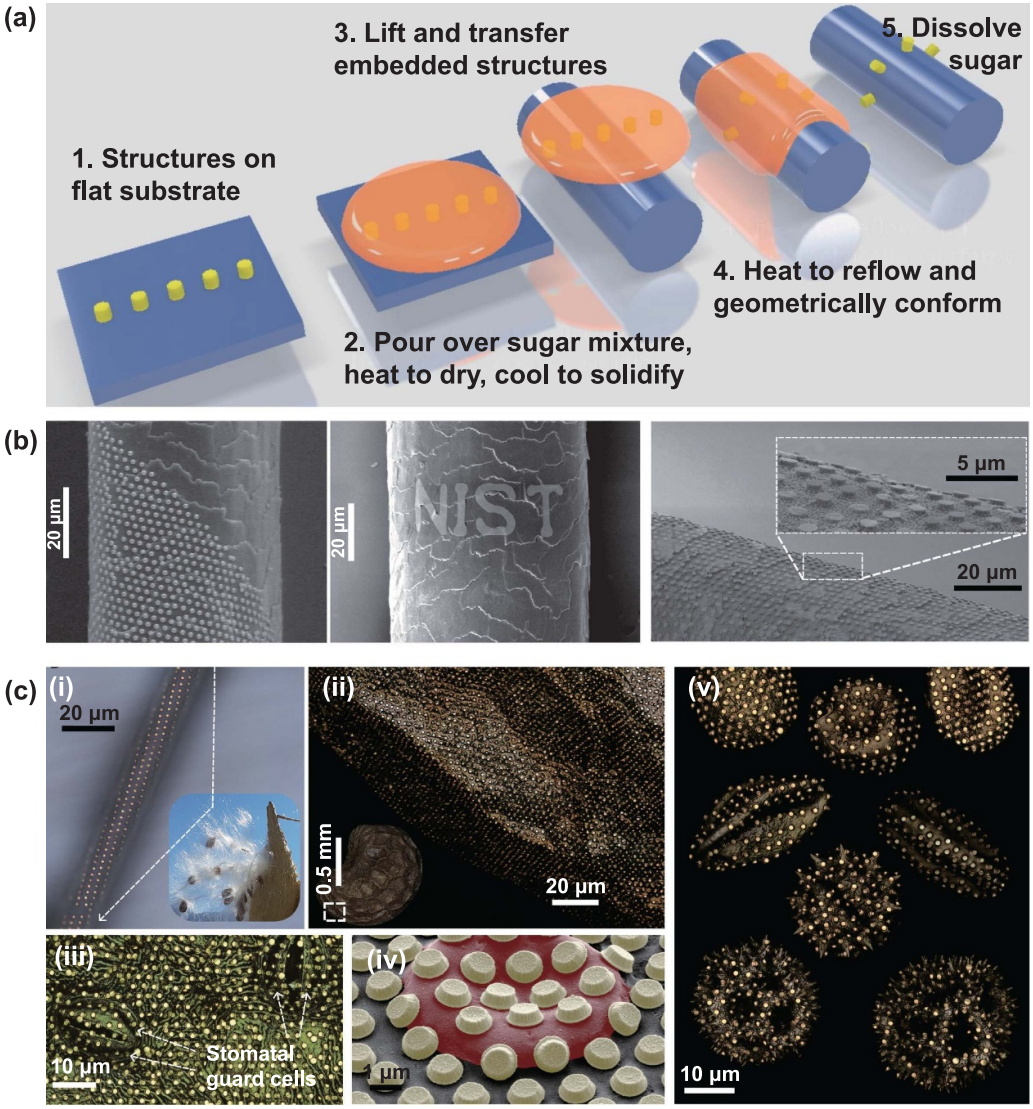

Figure 12. Reflow TP. (a) Schematic illustration of the process using reflowable sugar as a stamp. (b) Fabrication of isolated dots (left) and letter (middle) pattern on hair. Right: the enlarged SEM image of the hair surface. (c) Fabrication of Au disks on (i) floss fibres, (ii) poppy seed, (iii) leaf, (iv) red blood cell, and (v) pollen grains. (a)–(c) From [68]. Reprinted with permission from AAAS.

as metals, and the transfer of complete devices, the suitability and effectiveness of ice-assisted TP require more extensive research.

2.3.3. Reflowable material stamp. While conventional elastomer or flexible tape stamps can achieve high-precision transfer for most flexible substrates and materials, their deformation capabilities are limited, falling short of the requirements for high-curvature substrates or microstructures. Moreover, the pressure needed to lift devices and achieve conformal contact can potentially cause unpredictable damage to fragile devices or substrates. An alternative approach using a liquid medium or sacrificial layer as a stamp could circumvent these issues but at the expense of limiting the transfer accuracy of device placement [89, 135].

Recently, Zabow proposed a novel transfer stamp made from reflowable materials such as a sugar mixture, enabling high-conformal 3D TP [68]. The entire TP process using the reflowable sugar mixture stamp is depicted in figure 12(a). The process begins by pouring the sugar mixture onto a flat donor substrate with a pre-fabricated pattern. The mixture is then heated to evaporate any water and subsequently cooled to solidify at room temperature. This solidification allows the pattern to lift off from the donor substrate and become embedded in the sugar mixture layer. The receiving substrate is then coated with the sugar mixture layer containing the embedded pattern, which is heated to melt the sugar. Thanks to the reflow characteristics of the sugar, it can conformably adhere to a

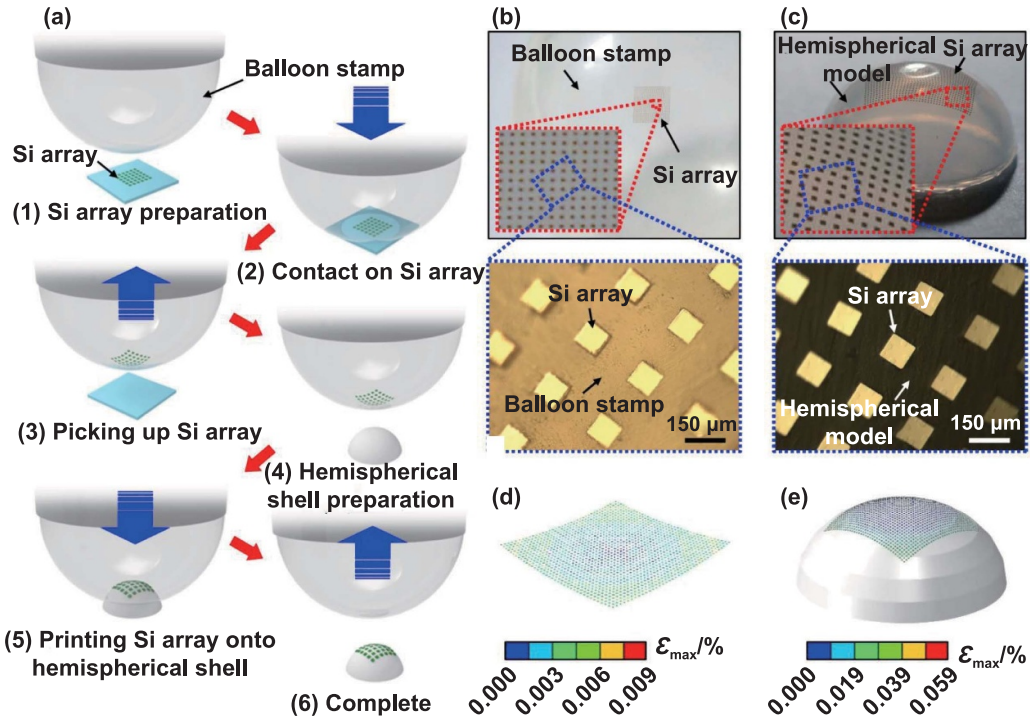

Figure 13. 3D TP using balloon stamp. (a) Schematic illustration of the TP process. (b) Digital and optical images of Si array on the balloon stamp surface. (c) Digital and optical images of the Si array printed on the hemispherical shell surface. (d) Maximum principal strain (εmax) contour of the Si array on the balloon stamp surface. (e) Maximum principal strain (εmax) contour of the Si array on the hemispherical shell surface. (a)–(e) Reproduced from [78], with permission from Springer Nature.

receiving substrate of any curvature, while preserving the relative position of the embedded pattern. The transparency of the sugar mixture also facilitates easy position alignment in subsequent steps. Lastly, the sugar layer can be directly delaminated or released by dissolving an additional sacrificial layer, such as a photoresist.

By leveraging this reflowable stamp, the author successfully achieved high-yield TP of nanoscale patterns onto various substrates with complex surfaces, even those with nanoscale radii of curvature, including metal, polymer, paper, hydrogel, hair, seeds and even cell surfaces (as shown in figures 12(b) and (c)). Furthermore, the inherent transparency of the sugar mixture provides a practical advantage for transfer alignment. While the potential applications of this property have yet to be fully explored, it holds promise for use in areas where high resolution is imperative, including optics, metasurfaces, and micro-LED technologies.

2.3.4. 3D curved stamp. To meet the demands of patterning on 3D curved structures, one effective approach is to design a conformable 3D curved stamp, such as a balloon surface [78]. Figure 13(a) illustrates the use of a balloon stamp to pick up a Si array from a donor substrate and transfer it to a hemispherical shell. The corresponding optical images are shown in figures 13(b) and (c). The conformal contact between the balloon stamp and substrates could greatly reduce the strain between the Si/stamp and Si/substrate interfaces. Based on FEA calculation, the strain in all the Si array (no matter center or edge) is less than 0.01% when peeled- off by balloon stamp, which is far less than the fracture strain of Si (figure 13(d)). The Si array could also maintain a minor deformation during the print process (figure 13(e)). The malleability of the inked balloon stamp outperforms that of conventional elastomer stamps, making it an ideal choice for a variety of 3D surfaces in applications such as antennas, smart contact lenses, and photodetectors.

Another innovative stamp design, inspired by the wrapper of a spherical candy, is depicted in figures 14(a)–(c) [69]. This stamp comprises an elastomer backed by a water-soluble tape. A custom-made prototype tool utilizes an air-induced pressure difference to achieve optimal conformability. The near-vacuum state causes the stamp to tightly wrap around the target substrate. Following UV exposure and curing of the pre-coated binder on the substrate surface, the target pattern is transferred to the 3D curved surface upon dissolution of the water-soluble tape.

The viability of this method for constructing 3D curved electronics has been demonstrated through the fabrication of spherical antennas, light-emitting diode arrays, and solar cells. This approach shows great promise for the advancement of 3D curved electronics manufacturing.

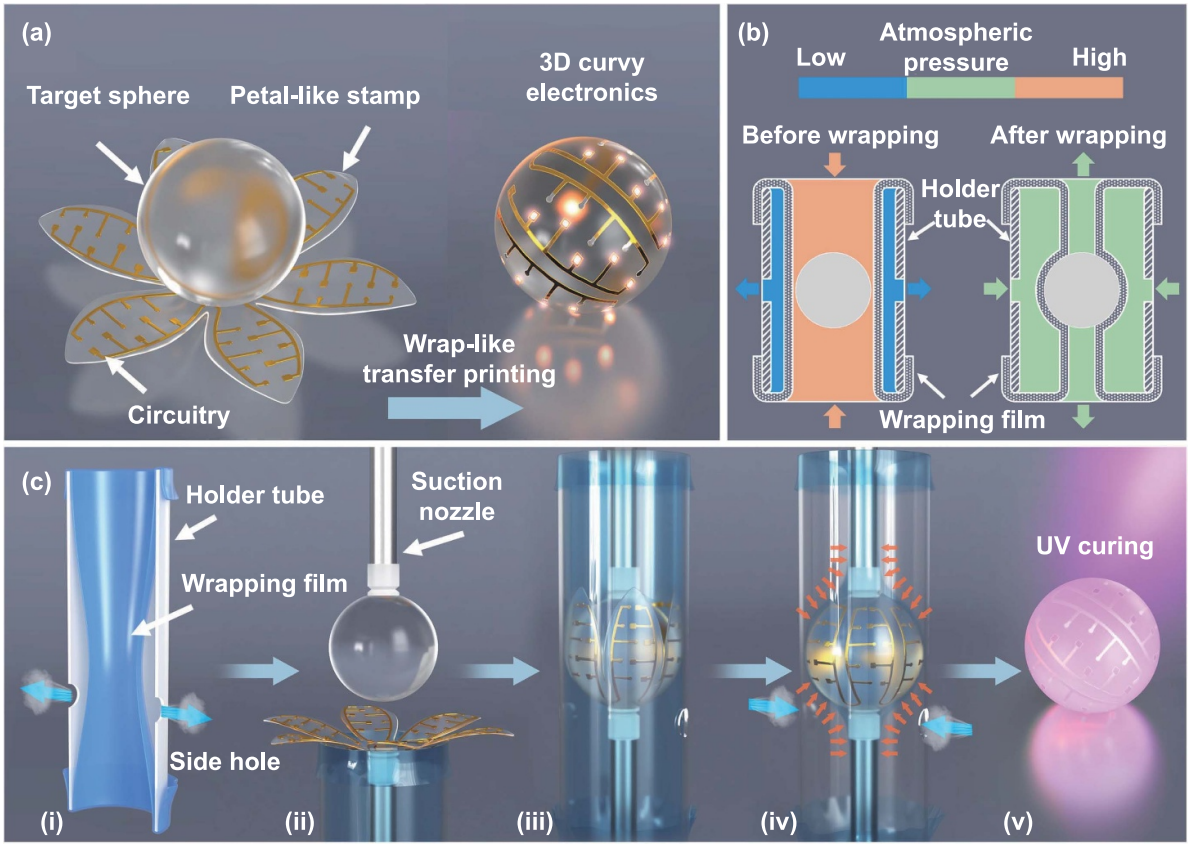

Figure 14. Wrap-like TP. (a) Structure of the petal-like stamp. (b) Pressure distribution before and after wrapping. (c) Schematic illustration of the TP process. (i) Inflate the stamp film. (ii) Alignment of the receiver substrate and stamp with the pattern. (iii) Insert the stamp and substrate into the inflated film. (iv) Contact and print of the pattern. (v) UV cure the binder between the pattern and substrate. (a)–(c) Reproduced from [69]. CC BY 4.0.

3. Conclusions and outlook

Over the past few decades, TP has emerged as a powerful and promising technique for fabricating a broad spectrum of flexible electronic and photonic devices. The success of device transfer is contingent upon the competing fracture at the substrate/device/stamp interfaces. To amplify the difference in adhesion forces between these interfaces, numerous modifications have been developed for the substrate/device interface during the pick-up process, such as water, graphene, self-assembled monolayers, and sacrificial layers. Similarly, modifications for the device/stamp interface during the print process include alterations to the contact area, magnetic or laser additions, and unique stamp designs like thermal tape, ice, sugar, or 3D curved stamps. These advanced TP methods can accommodate nearly all material types, including metals, semiconductors, polymers, and receiver substrates, ranging from rigid, flexible, and stretchable to curved and complex 3D surfaces.

The future of TP hinges on exploring wider application possibilities. Depending on specific application scenarios and device preparation processes, it is vital to select the appropriate stamp and modification method to facilitate the non-destructive transfer of devices. On one hand, the mastery of interfacial interactions is crucial for ensuring high-quality transfer during the fabrication of flexible ICs, two-dimensional materials, and semiconductors for applications such as diodes. Control over these interfaces directly impacts device performance and integrity. On the other hand, the bespoke design of stamps plays a pivotal role in broadening the selection of substrates and facilitating the assembly of intricate structures. This level of customization is particularly vital in fields such as micro-nano optics and biomedicine, where the complexity of structures often dictates functionality. We anticipate that the researchers could reference this review to find suitable methods when facing different challenges in device manufacturing. Despite the success rate of transfer, it is also significant to consider the balance between resolution, throughput and cost of the printing methods in production. This is an indispensable step towards industrialisation. Other challenges such as solvent compatibility, temperature sensitivity, and a universal way to realize the high precision alignment are worth deep studying in the future. TP is anticipated to play a crucial role in the realms of wearable electronics and biomedical applications. It holds great promise for further advancements in these rapidly evolving fields.

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Conflict of interest

The authors declare no competing interests.

ORCID iD

Zijian Zheng https://orcid.org/0000-0002-6653-7594

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