INVITED PAPER

Inorganic Materials and Assembly Techniques for Flexible and Stretchable Electronics

Important progress has been made in developing design strategies, materials, and associated assembly techniques that provide approaches to electronics with unconventional formats.

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ABSTRACT | In recent years, important progress has been made in developing design strategies, materials, and associated assembly techniques that provide empowering approaches to electronics with unconventional formats, ones that allow useful but previously hard to realize attributes of function. Notable examples of the progress made include: light weight, large area, high performance electronics, optics, and photonics; electronic and optical systems with curvilinear shapes and capacities for accommodating demanding forms of mechanical flexure; new device form factors for use in sensing and imaging; the integration of high performance electronics in 3-D with demanding nanometer design rules; functional bioresponsive electronics; and advanced hybrid materials systems for lighting, energy storage, and photovoltaic energy conversion. In this report we highlight advances that are enabling such promising capabilities in technology—specifically, the fabrication of device elements using high performance inorganic electronic materials joined with printing and transfer methods to effect their integration within functional modules. We emphasize in this review considerations of the design strategies and assembly techniques that, when taken together, circumvent limitations imposed by approaches that integrate circuit elements within compact, rigid, and essentially planar form factor devices, and provide a transformational set of capabilities for high performance flexible/stretchable electronics.

KEYWORDS | Bio-integrated electronics; flexible electronics; inorganic materials; integrated systems; stretchable electronics; transfer printing

I. INTRODUCTION

The fabrication of high performance integrated circuits provides examples of the most sophisticated manufacturing methods, as well as the most high-performance materials, used in any area of modern technology. The advanced functional systems they provide are ones that are generally characterized by a massive integration of circuit elements within compact, rigid and essentially planar form factor devices. The models of biology offer interesting points of comparison—ones where system elements are integrated with challenging 3-D design rules embedding materials structures that are frequently soft, curvilinear, and highly flexural in their functional form. It represents an interesting challenge in technology to develop strategies through which existing classes of electronic materials (EMs)—ones affording capabilities for high-circuit-level performance—might be utilized in conceptually related ways to provide new functional capabilities. The envisioned uses include electronic technologies permitting intimate integration with biological tissues, new capabilities for measurement and sensing, efficient low-cost systems for energy transformation and storage, and perhaps most importantly modes

Manuscript received October 18, 2014; revised December 18, 2014; accepted January 18, 2015. Date of current version May 19, 2015.

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Digital Object Identifier: 10.1109/JPROC.2015.2396991

of large-area-integration for applications not countenanced by Moore's law [1]. Exemplary cases here include: electronic skins [2] and medical devices [3] that provide advanced human-machine interfaces; bio-inspired cameras [5], [6] embedding nonplanar/curvilinear electrooptics; wearable electronic systems [7]; deformable displays [8]; and conformal mechanical energy harvesters [9]. One critical requirement for all technologies of this type is the ability to accommodate extremes in mechanics that specific applications engender while preserving the performance capabilities of their electronic systems.

Organic materials (OMs) figured centrally in many of the earliest explored approaches to flexible/stretchable electronics. It has long been realized that organic semiconductors, many of which are intrinsically flexible, might well serve as the active components of devices. The exemplary use of both molecular and polymeric semiconductors in devices such as organic light-emitting diodes (LEDs) [10], [11], organic solar cells [12], [13] and organic field effect transistors (FETs) [14] illustrate their potential for use more broadly in flexible/stretchable electronics. Even so, OMs suffer from the very nature of their charge transport mechanisms, which generally render them unsuitable for use in applications requiring high performance electronics. It remains a fact that, from a device-level perspective, established inorganic materials in thin-film, single-crystalline forms (e.g., GaN for LEDs [15], GaAs for solar cells [16], and Si for metal oxide semiconductor field effect transistors (MOSFETs) [17]) exceed the levels of performance that can be realized using thin-film organic semiconductors. This presents an interesting opportunity for current research to provide means through which generally brittle inorganic electronic materials can be integrated in thin film forms appropriate for use in flexible and stretchable electronics.

An empowering approach to this problem is found in emerging design concepts that use optimized material configurations and structural layouts on soft, flexible, or elastomeric thin film substrates. To introduce properties of flexure, bulk inorganic materials are replaced by 1-D or 2-D nanoscale counterparts that are very thin—a feature that limits the magnitude of the strains occurring during deformation. This approach has been demonstrated with diverse classes of materials, with Ag nanowires [18], and carbon nanotubes [19], silicon nanoribbons [20], and InP nanomembranes [21] serving as exemplary cases. Form factors that are stretchable constitute a more challenging characteristic of mechanics to address. To do so, the integration of an inorganic device element must be done in ways (e.g., "wavy" and 3-D configurations, etc.) that allow out-of-plane motions to decouple the active electronic components from large amplitude strains that would be sufficient to induce mechanics-based materials failures. Taken together, these emerging concepts demonstrate generalizable designs for stretchable/flexible electronic systems offering circuit performance characteristics comparable to on-wafer/chip-level counterparts, yet having capacities for mechanics comparable to those of biological tissues or rubber bands.

The development of any new form factor for electronics technology carries with it a correlated challenge of establishing efficient, low-cost, high-yield means for their fabrication. Towards this end, printing and transfer-assembly techniques [22] have come to provide an increasingly attractive manufacturing approach for stretchable/flexible electronics. Established device designs and processing approaches can be adapted to such modes of manufacturing. EMs of diverse form (e.g., wafer-derived silicon nanoribbon/nanomenbranes [23] and vapor-liquid-solid-grown nanowires [24]) or even fully integrated devices can be prepared on a source substrates and printing/transfer-assembly methods used to deliver and assemble them on mechanically-compliant substrates that can flex or stretch. To be useful, these methods must meet stringent requirements for spatial precision, registration, yield, and throughput, among others.

In this review, we describe recent advances that have been made in design concepts, fabrication methods, and assembly techniques—ones exploiting high performance inorganic EMs—as a foundational approach to flexible and stretchable electronics. Several exemplary applications are discussed that highlight the advanced functionalities that can be realized. We first describe the materials and mechanical requirements for the use of inorganic EMs in flexible and stretchable forms. We follow with a survey of the printing/assembly techniques that enable their use, highlighting examples of printable inks embedding fully functional forms of electronic integration. In the sections that follow, we present exemplary cases where these materials sets, and their associated means of processing and fabrication, have been used to illustrate both new functional capabilities and realistic pathways to manufacturing that would allow their use in technology.

II. MATERIALS AND MECHANICS

A. Inorganic Materials for High Performance Flexible Electronics

The most elementary strategy for enabling flexibility in "rigid," hard inorganic materials involves a simple idea of mechanics—for a given deformation, strain decreases linearly with decreasing materials "thickness." For a thin film, this relationship is described by $\varepsilon = t/2r$, where ε is the strain, t is the thickness of thin film and r is the bending radius [25]. For example, this relationship predicts that a silicon ribbon 100 nm thick can bend to a radius of 7 μm before cracking at a limiting strain of 0.7% [26]. A local bending radius of $\sim$340 nm, for example, was shown experimentally for silicon nanowires with a radius of 39.2 nm [27]. This is a completely general idea—any material can flex as long as it is sufficiently "thin."

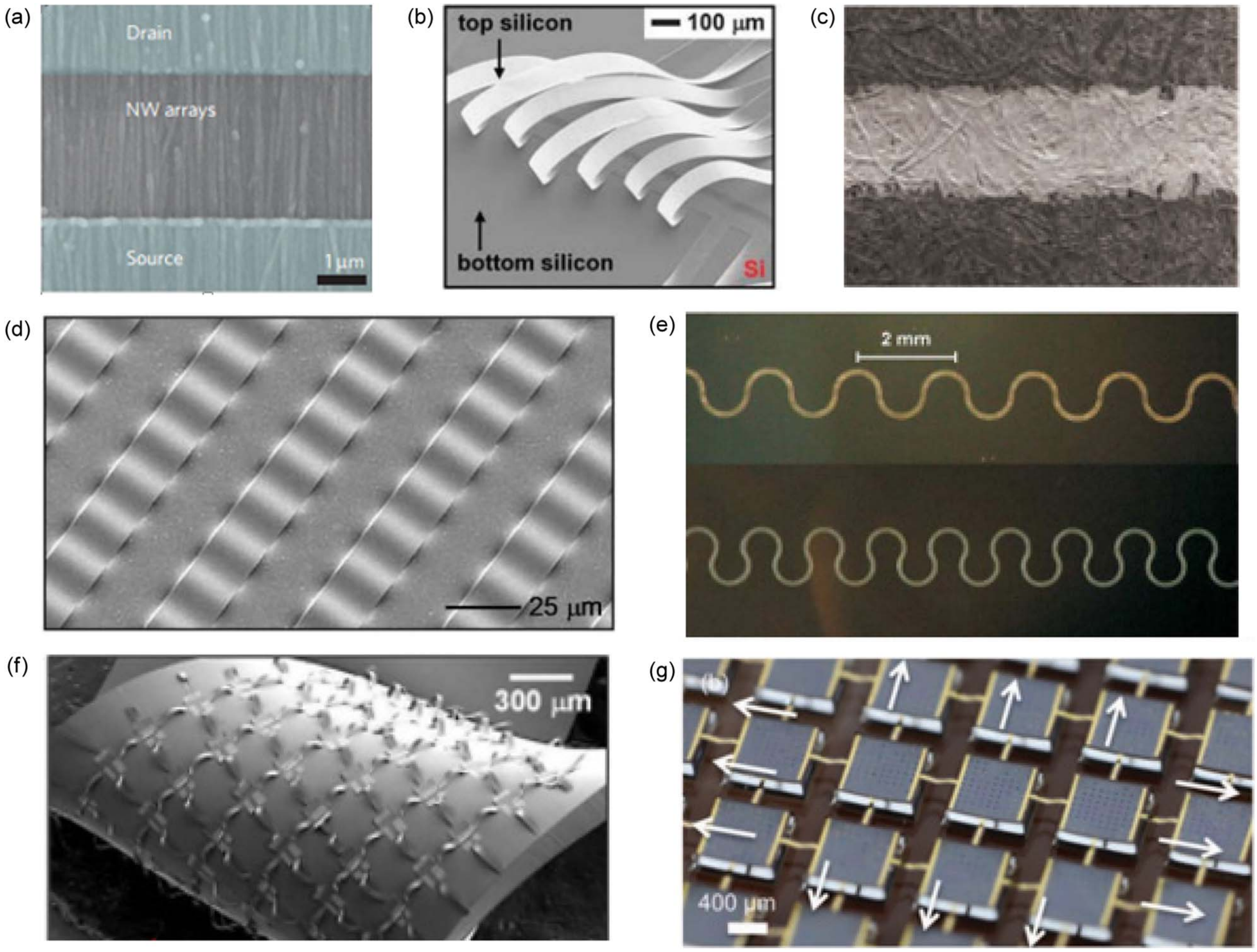

Fig. 1. ***Material configurations and structural layouts for flexible/stretchable electronics. (a) Aligned arrays of 1-D Ge/Si core/shell nanowire arrays, here used as the channel materials for active-matrix FETs supported on a polyimide substrate. Reproduced with permission from [28]. Copyright American Chemical Society. (b) Patterned 2-D silicon nanoribbons undercut and delaminated from a Silicon-on-Insulator (SOI) wafer using an isotropic wet-chemical etch of the buried oxide layer. Reproduced with permission from [35]. Copyright The American Association for the Advancement of Science. (c) Ag nanoparticles patterned by direct-ink writing on a paper substrate. Reproduced with permission from [41]. Copyright John Wiley and Sons. (d) Buckled layout of printed silicon ribbons upon release of prestrain in the elastomeric (PDMS) substrate. Reproduced with permission from [35]. Copyright The American Association for the Advancement of Science. (e) Planar Au filamentary serpentine interconnects on an elastomeric substrate before (upper panel) and after (lower panel) compression. Reproduced with permission from [48]. Copyright IEEE. (f) Out-of-plane bridge interconnects for semiconductor circuits illustrating 3-D structural motifs formed by strain-minimizing release dynamics Reproduced with permission from [51]. Copyright National Academy of Sciences, USA. (g) High performance GaAs solar microcells interconnected by filamentary gold wires patterned in an open mesh layout to accommodate large amplitude strain evolution in the supporting substrate. Reproduced with permission from [53]. Copyright John Wiley and Sons.***

For devices using 1-D EMs (for example, in the form of nanotubes and nanowires), it is possible to accommodate an applied strain while maintaining pathways for charge transport and, with proper layout, large amplitude strains that minimally perturb electronic properties can be realized. Fig. 1(a) illustrates one example, here aligned Ge/Si core/shell nanowire arrays which are used as the channel material for field effect transistors (FETs) supported on a flexible polyimide (PI) substrate. These arrays showed a modest decrease in conductance ($\sim$6%) when bent to a radius of 2.5 mm along the axial direction [28]. In a similar fashion, single-walled carbon nanotubes supported on polyethylene terephthalate (PET) maintain high conductance even at a bending radius of 1 mm owing to their small diameter (1–2 nm) [29]. In some cases, tensile strains can be accommodated when the nanowires are deposited on a deformable substrate in randomly oriented, multilayer forms. For example, mesh-like 500 μm long Ag nanowire networks showed high conductance with excellent mechanical compliance to strains reaching to 460% (defined by the substrate) [30]. Bundles of SWNTs (lengths > 1 mm) also provide printable routes to stretchable conductors, as illustrated in recent report that dispersed the nanomaterial conductors in a rubber matrix to form a gel-like conductive ink [31].

Similar forms of flexure can be realized using 2-D EMs, again with the proviso that they are thin. Systems of this form, as exemplified by semiconductor nanomembranes (NMs) and nanoribbons, have been intensively studied and important advances towards mechanically robust systems reported. For example, a fully functional cardio electrophysiology sensor system integrating 2016 silicon NM transistors on a plastic sheet allowed flexure to radii of curvature as small as 500 μm without loss of performance [32]. A notable benefit attending the use of NM materials is that they can be taken directly from high-purity single-crystal wafers with proper release mechanisms and integrated NM devices fabricated using well established means of wafer scale processing [33]. For example, with suitable alignment of trench cuts, silicon NM platelets and ribbons can be released from a (111) silicon wafer by an anisotropic wet etch with KOH; this is a procedure exploiting highly anisotropic etching characteristics of low index Si planes [34]. The image in Fig. 1(b) shows a related anisotropic wet-etching fabrication method, here the buried oxide layer of a silicon-on-insulator (SOI) wafer etched using HF to detach silicon ribbons defined by a prior RIE trench cut [35]. An intriguing example reported in the recent literature used epitaxially grown multilayer stacks of GaAs thin film photovoltaic cells interspaced by layers of AlAs. The selective etching of the AlAs by HF allowed a quantitative release of the high-efficiency GaAs PV microcells and recovery of the growth substrate [36]. This latter example illustrates the utility of NM materials for use in cases where high performance electronics are required to meet application needs. Specific examples of the latter are given in the sections that follow.

Nanoparticles also comprise a system of great interest for use as functional materials in flexible and stretchable electronic devices. Metal nanoparticles in fluid suspension, for example, constitute a useful ink for printing metal electrodes and bus-level interconnects [37], which, after further processing by annealing [38] or laser sintering [39] yield highly-conductive, and with proper design, flexible metallic structures. An example of note was reported for a silver nanoparticle ink [40] that was used to print Ag structures on a number of challenging substrates, including paper and polyimide. The image in Fig. 1(c) shows a section of a large area array of Ag conductors printed on paper, structures which retained high conductivity to a bending radius of 9 mm [41].

B. Form Factors for Extreme Flexure and Stretchability

There has now been substantial progress made in devising form factors for electronics using inorganic EMs in nanoscale form that can accommodate extremes in mechanics, including previously unprecedented capabilities for flexure and stretching. The most effective design approach for electronics that can accommodate large amplitude effective strains without fracture are based on various forms of curvilinear/3-D integration of the electronic device elements. Wavy structures of single-crystalline Si, for example, can be fabricated by printing thin film ribbons on a stretched elastomeric substrates. The release of the prestrain leads to a buckling instability that forces the film into a wavy configuration [42], [43], as illustrated by image shown in Fig. 1(d). This allows an evolution of the strain in the thin film during elongation (relaxation) that is much smaller in magnitude than that experienced by the substrate [44]. It was found, for example, that 100 nm thick single crystal silicon ribbons bonded on PDMS, similar to the ones in Fig. 1(d), experience a peak strain during tensile deformation that is 15 times less than that of the underlying PDMS substrate [45].

A conceptually related example is based on a 2-D curvilinear form. For this case, filamentary wires are patterned as reiterative circular arcs that allow large tensile deformations with minimum stress concentration [46]. The strain mitigation for structures of this type results due to both elongation and some degree of out-of-plane deflection [47]. The Au wires shown in the Fig. 1(e) illustrates the motif as applied to an array of serpentine interconnects [48]. These specific structures were found to be mechanically robust—withstanding large magnitude bending, stretching, compressive, and twisting deformations without fracturing. Computational models allow assessments of the width and thickness, radius of curvature, and spacing of the metal lines to optimize electrical and mechanical performances [49]. Complex, open mesh configurations, such as fractal designs [50], have also been proposed as a means to accommodate more extreme forms of elastic deformation. Perhaps the most interesting strategies for enabling extreme forms of mechanics are ones based on intrinsically 3-D forms of integration of the elementary inorganic EM structures. This is illustrated by the example shown in Fig. 1(f), where an interconnected square-mesh of thin-film Si wires was bonded only at the nodes to a prestrained elastomeric substrate. Release of the strain leads to a popped-up motif of the interconnecting wires which in turn serves a deformable bridge architecture [51]. The resulting interconnecting "bridges" can freely move out of plane as shown in Fig. 1(f) and in this way provide a robust means through which to accommodate 3-D forms of large amplitude mechanical deformation.

The latter in-plane or out-of-plane designs appear to be especially important as they allow the fabrication of complex electronic systems as arrays in which highly deformable/stretchable conductors can be used to interconnect active circuit components at nodes that experience low strain. An exemplary case is illustrated by the structures formed by 300 nm thick CMOS ribbons bridged by polymer-embedded metal interconnection lines. When printed on a prestrained PDMS surface [52], the wires buckle vertically off the PDMS after release of the strain—forming a highly elastic noncoplanar mesh of bus interconnects. This structure can be compressed, stretched, twisted and sheared at a degree depending on the stiffness and length of the bridges. Another intriguing example is illustrated in Fig. 1(g), which shows 3.6 μm thick GaAs solar microcells printed on textured PDMS islands with each pixel separated by trenches and bridged by filamentary gold interconnects. The maximum shear stress for this layout is eight times less than that for a flat substrate [53].

Finally, we note that the failure of a circuit via a mechanics-induced fracture can be mitigated by isolating components from the strain by adopting a neutral-mechanical-plane design. To do so, a device element is placed within a thin film at a location where strain is minimal for an arbitrary bending radius [52]. This is most commonly done by coating the assembly with an additional (e.g., polymeric) encapsulation layer [54]. This is a multifunctional aspect of integration in that it can serve as a means of both mechanical and environmental protection.

III. FABRICATION: PRINTING AND TRANSFER ASSEMBLY METHODS

Any commercially viable flexible/stretchable electronics will require reliable, high-throughput means of manufacturing. Owing to the small dimensions and mechanical fragility of nanoscale inorganic EMs, their assembly and functional integration with high spatial and orientational precision remains an important challenge towards which considerable effort has been directed in research. Methods exploiting self-assembly, in which EM structures and/or device level components are placed in fluid suspensions and deposited on a substrate by electrical/magnetic forces [55], [56], flow fields, [57], [58], or recognition by surface relief structures [37], have been intensively studied. Although they show promise, current capabilities remain less than those required for commercial applications. Printing techniques, of which many forms have been investigated, constitute important and highly promising alternatives. We highlight three exemplary cases that illustrate the potential for printing (and the highly related technique of transfer assembly) as approaches to manufacturing for flexible/stretchable electronics.

A. Printing and Transfer Assembly

Wet printing techniques, such as screen printing [59], flexographic printing [60], and inkjet printing [61], to name a few, have proven to be extremely useful in the fabrication and assembly of organic electronics with applications ranging from organic LEDs [62], [63] to organic solar cells [12], [64]. These methods, though, are mostly restricted to fluid inks of predefined requirements for viscosity and homogeneity that limit their use with inorganic EMs. One notable exception is found in printing nanoparticle inks in a highly deterministic manner. For example, direct ink writing represents an attractive approach, one offering capabilities for patterning challenging geometries embedding both 2-D and 3-D design rules [65], [66]. It is an additive method of fabrication and in this regard offers useful attributes that might complement more conventional means of patterning. The images in Fig. 2(a) illustrate the method and exemplary applications of direct ink writing, here applied to metallic interconnects [40]. A print head with a fine tapered cylindrical nozzle is used to deliver the ink in patterns that are determined by the diameter of the nozzle, rheology of the ink, and movements provided by high-resolution translation stages. A prototypical ink in this case might consist of metallic or semiconducting nanoparticles in a fluid suspension whose detailed composition is engineered to optimize the patterning and electrical/structural performance of high resolution material structures. This method can be used to print complex, and even free-standing structures, examples of which are shown in the lower two insets of Fig. 2(a) for metallic Ag conductors. After annealing, the Ag features exhibit high conductivity and excellent capacities for flexure [as illustrated by the spanning 3-D wires printed on a supporting metal spring (lower panel)].

Contact printing and transfer assembly frame complementary methods that can be used to pattern essentially any form of inorganic EM in the form of a solid ink, with exemplary demonstrations reported in the literature for cases that include nanowires [28], nanotubes [67] and nanomembranes [68]. The patterning in this case results from a physical transfer of material (a "solid ink") from a growth/preparative substrate to a receiving one. The transfer process is typically enabled by a difference in adhesion forces (exploiting both dynamical and chemical means of control). The images in Fig. 2(b) illustrate show one example, here the transfer of aligned Ge nanowires to a semiconductor substrate [69]. In this process case, the nanowires were grown on a cylindrical roller substrate using a VLS growth method. The movement of the roller over the substrate transfers the nanowires in an aligned fashion. The highly aligned nanowires were subsequently processed in functional device forms that included field-effect transistors and photodetectors. It is believed that methods such as this can be easily scaled for use in the fabrication of large area flexible electronics. Challenging issues of materials selection, testing, feature size and spacing, and property control remain to be addressed, however.

Over the last decade, various forms of transfer printing using an elastomeric stamp as a printing tool have attracted great attention, in part due to the diversity of the print inks that can be patterned in high yield and high spatial uniformity [22]. The generic process is shown in the upper panel of Fig. 2(c). In the inking step, a stamp is placed in contact with a donor substrate where, upon peeling, the ink is released. In the printing step, a contact made to the receiving substrate leads to the release and transfer of the ink [70]. Many methods have been developed to control the aspects of this sequence of mass transfer steps—methods based variously on the modification of surface, interfacial, and bulk materials properties of the

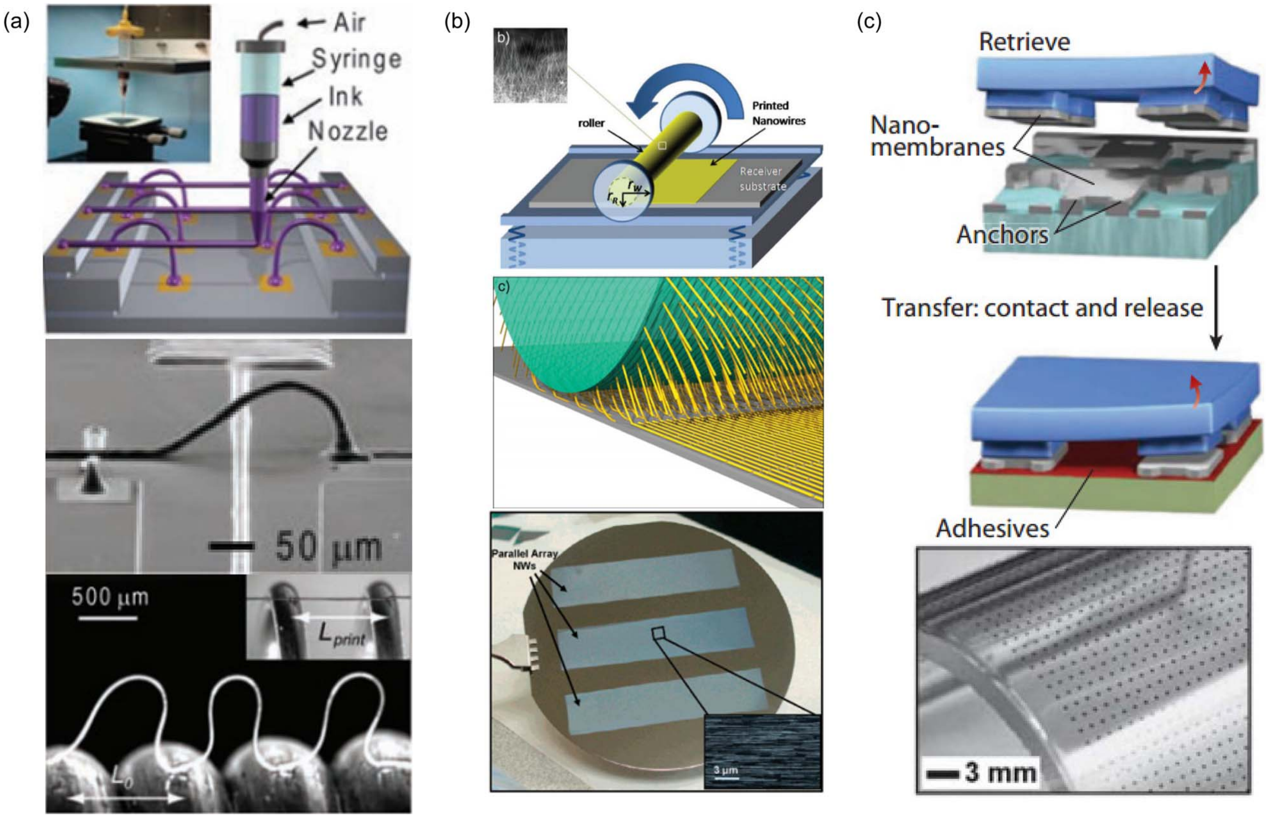

Fig. 2. ***Printing techniques with important applications in stretchable/flexible electronics. (a) A schematic depiction of direct ink writing (upper panel); an exemplary silver interconnect in the form of self-supported arch printed over an intersecting bus line (middle panel); self-supporting Ag wires printed on a compressed spring (lower panel), the inset shows the wires after extension on the spring. Reproduced with permission from [40]. Copyright The American Association for the Advancement of Science. (b) Schematic depiction of a roller-based contact printing apparatus for nanowire arrays (upper panel) and roll-to-roll printing of nanowires grown by a VLS process on the roller printing head (middle panel); Aligned Ge nanowire arrays printed on a 4 in Si/SiO_2 wafer (lower panel). Reproduced with permission from [69]. Copyright John Wiley and Sons. (c) Schematic depiction of transfer printing of a solid ink using an elastomeric stamp as a printing tool showing the inking and transfer stages of the process (upper and middle panels); exemplary large area arrays of single-crystalline GaAs micro-chiplet inks printed on a curvilinear surface (lower panel). Reproduced with permission from [70]. Copyright Nature Publishing Group.***

stamp/substrate pairings as well as the dynamics of the adhesive forces. For the former, useful means include laser-induced thermo-expansion [71], adhesion modification layers [72], plasma activation [73], [74], and shape engineering of the stamp contact interface [75], [76]. For the latter case, detailed mechanics models of the transfer processes [77] illustrate the essential role played by dynamics such as shear [78] and/or peeling speed [79] in controlling the attributes of mass transfer. The span of the design rules that can be accommodated are vast—literally spanning from nanometers to many centimeters [80] depending on the attributes of the stamps employed. Aided by automated tool sets [81], printing accuracies of better than 200 nm [82] with a throughput of over one million pixel elements per hour [80], [83] have been demonstrated, along with capabilities for precise control over areal densities and stacking configurations [84]. The image in the lower panel of Fig. 2(c) presents an example illustrating such operative capabilities, a section of a large area array of GaAs platelets printed on a curvilinear substrate. A particular advantage of the transfer-assembly method demonstrated here is that it is a highly parallel method of printing that also remarkably broadens the materials choices for inks by separating all aspects of materials growth and preprint fabrication/processing from the inking and printing steps. It also preserves the electronic properties of such generally fragile nanomembrane/thin-film inks due to the soft nature of the stamps and the generally non-destructive forces that mediate the mass transfer. Given these potential advantages, transfer printing stands almost alone in its capacity to enable the high-throughput assembly of sophisticated/fully-functional circuit-level components—an essential requirement for integrating advanced functionalities (such as logic operation) into flexible/stretchable electronics. In Section III-B, we highlight several examples of functional, full device-level inks of this type.

B. Functional Electronic Inks for Flexible and Stretchable Electronics

Inks arguably constitute the most important enabling component of flexible/stretchable electronics enabled by transfer assembly means of fabrication. They define such features as the circuit-level functionality of the device, its electronic performance attributes, and capabilities for enabled applications. A large range of materials have been prepared in forms that render them suitable for use as printable inks for use in flexible and stretchable electronics and other devices, including many examples of inorganic semiconductors (e.g., as nanomembranes, nanoribbons, nanowires, quantum dots [85], [86]), metallic nanoparticles [87], [88], organic materials (e.g., polymeric resists [89], [90] and even living cells [91], [92]). Low dimensional, essentially 1-D materials, such as nanowires and nanotubes, are particularly attractive materials for use as components in high performance devices because of their bendable structure and high electron mobility. Indeed, the advantages afforded by these enabling attributes have been demonstrated in a number of examples reported in the recent literature. Notable examples include flexible optoelectronic [93] and mechanical sensing devices [94] fabricated using such active material elements. Even so, the fabrication of complex flexible electronic systems using semiconductor nanowire arrays as a start remains limited by numerous challenges that have yet to be fully addressed in research. Of these, the typically poor thermal and chemical compatibilities of flexible substrate materials with the harsh conditions required for post-print processing and patterning of fully integrated electronic components are particularly limiting. In addition, high degrees of precision in layout and multilevel registration are difficult to obtain on substrates other than hard, planar handling/source material wafers, which, because of error propagation, limits the permitted levels of integration and thus performance of the resulting circuit.

Fabrication methods based on transfer assembly, when conjoined with 2-D EM inks derived from high-purity single-crystalline wafers, can circumvent many of the aforementioned limitations, not only because of their superior electronic properties but also due to their planar geometry—one that is most compatible with both the established tools and processes of microelectronics fabrication. The inks in this case are fully fabricated as a functional 2-D circuit element with all the required processing steps carried out on an electronic grade semiconductor source wafer using established procedures. The completed devices are then released from the source wafer and printed on a receiving substrate using a scalable/highly-parallel transfer-assembly method. The section that follows discusses several exemplary cases in which sophisticated/highly-functional electronic inks are fabricated and printed in this way—benchmarks establishing the enabling promise of such materials for use in flexible/stretchable electronics.

The image in the upper panel of Fig. 3(a) shows small, micron-scale InGaN LEDs after completion of their fabrication and preparation for release from their material-source wafer [95]. The MBE-grown multilayer stack supported on a Si(111) base substrate was processed to hold the undercut LEDs in place via a system of anchors that are sacrificed during the printing step. Standard techniques employed to fabricate the LED inks include photolithographic patterning, epitaxial growth, reactive ion etching, contact metallurgy, thermal annealing, and anisotropic wet chemical etching (of a sacrificial layer to effect the undercutting). The resulting micro-LED arrays developed on the wafer in this way are fully functional, with n- and p-type contact pads ready for interconnection. Deterministic printing via a soft elastomeric stamp yields a nondestructive and highly parallel transfer. This is illustrated by the image shown in the middle panel of Fig. 3(a), where the emission of a blue-emitting array of LEDs printed on a polyimide substrate is shown after electrical interconnection. The resulting LED devices show virtually the same emission spectra and current-voltage characteristics before and after the printing step, as is illustrated by the data shown in the lower panel of Fig. 3(a).

As a second example, the upper panel of Fig. 3(b) shows an array of silicon solar micro-cells embedded in a luminescent solar concentrator waveguide by transfer printing [96]. The individual cells in this case are 15 μm thick, 50 μm wide and several millimeters long. The cells are processed prior to printing to enable high photon conversion efficiencies to electrical power. A crucial sequence of doping steps is needed to this end that carries a very high thermal budget, a feature enabled by the fact that all the microcell processing steps are carried out on the Si source wafer prior to printing. One notes that even with the high aspect ratio of the embedded silicon micro-cells shown, the printing is able to maintain the orientation and spacing with high precision, thus allowing optimization of the array to maximize light collection and conversion to electrical energy. Encapsulation of the solar micro-cell arrays yields a neutral mechanical plane design that shows both mechanical and electronic robustness to bending radii reaching to 4.5 mm [lower panel of Fig. 3(b)].

As noted above, inks requiring extreme physicochemical conditions during processing (e.g., metallization, doping, contact annealing, thermal oxidation, etc.) can be developed on their material-source wafers, printed on a common substrate, and after interconnection provide system-level functionalities that can greatly enhance the performance attributes of flexible/stretchable electronics. One notable example is shown in Fig. 3(c), where the circuit-level integration of high performance silicon MOSFET devices on a PI substrate are illustrated [97]. The individual silicon MOSFET devices were fabricated on a SOI wafer using process steps that included solid-state doping (1000 °C), thermal oxidation (1100 °C), RIE, wet chemical etching, and PECVD processing. The devices are

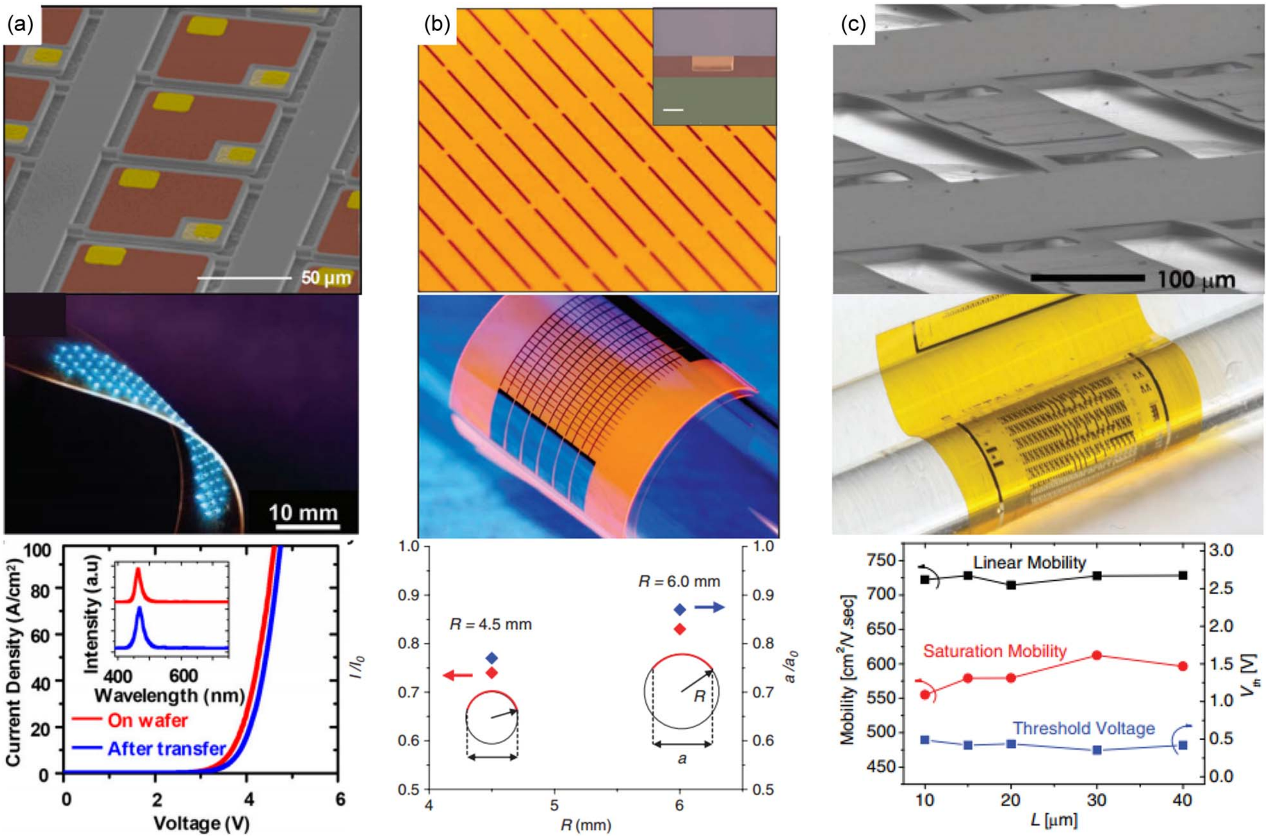

Fig. 3. ***(a) Micro-sized, blue emitting InGaN LEDs fabricated and supported on a mother wafer in a form suitable for use as a solid ink for functional patterning using contact printing/transfer assembly methods (upper panel); An array of blue-emitting micro-LEDs printed and interconnected on a supporting flexible substrate (middle panel), the small LEDs in this example contain an amount of semiconductor material that is approximately the same as is found in one standard commercial LED; Current density-voltage curves and emission spectra before and after transfer printing, showing only negligible performance differences (lower panel). Reproduced with permission from [95]. Copyright National Academy of Sciences, USA. (b) Silicon solar microcells embedded as an array in a flexible luminescent solar concentrator (upper and middle panels). Data for the photocurrents measured in different bending states showing that the electrical performance is not adversely impacted by mechanical flexure, here ratio of short circuit currents (I/I_0) scaling in a manner that is proportional to ratio of projection area (a/a_0) under normal incident illumination (lower panel). Reproduced with permission from [96]. Copyright Nature Publishing Group. (c) Silicon-silicon dioxide MOSFETs supported on wafer after fabrication prior to printing (upper panel); The same transistors after printing on a flexible polyimide (PI) substrate (middle panel); Threshold voltage and mobility values for the MOSFETs plotted as a function of the channel length as measured on the PI substrate (lower panel). Reproduced with permission from [97]. Copyright John Wiley and Sons.***

shown supported by their anchors on the source wafer after undercutting in the upper panel of Fig. 3(c). The devices were then printed onto a PI substrate, followed by metal interconnection (middle panel). Electrical data measured on the flexible substrate show sharp ON-OFF transitions, high field effect mobilities of 710 $cm^2 v^{-1} s^{-1}$, and subthreshold swings of 0.18 V $decade^{-1}$, all indicating high performance levels for the MOSFETs. Functional integrated circuits, such as inverters, NOR and NAND logic gates, were demonstrated as a proof of concept of the computing capability for flexible electronics of this form.

IV. INTEGRATED SYSTEMS

Integrated flexible/stretchable electronic systems promise system-level characteristics beyond what conventional on-wafer layouts can provide. For example, a traditional emissive display exploits fixed pixel arrays supported on a rigid glass panel. Printing methods provide interesting alternatives to the integration strategies used in such displays, with attendant consequences for both weight and mechanical durability [41]. Illuminating elements can be affixed and connected in a desired manner on a flexible substrate using only printing methods. An example of such is seen in Fig. 4(a), where an interesting lighting effect of a single LED chip mounted on paper is created using a hand-written conductive graphical trace. Fig. 4(b) shows a much larger array of LEDs chips supported and similarly interconnected on paper; the resulting module sustains high degrees of curvilinear deformation without engendering critical failures of the electrical components. Another intriguing example is shown in Fig. 4(c), an image showing a so-called

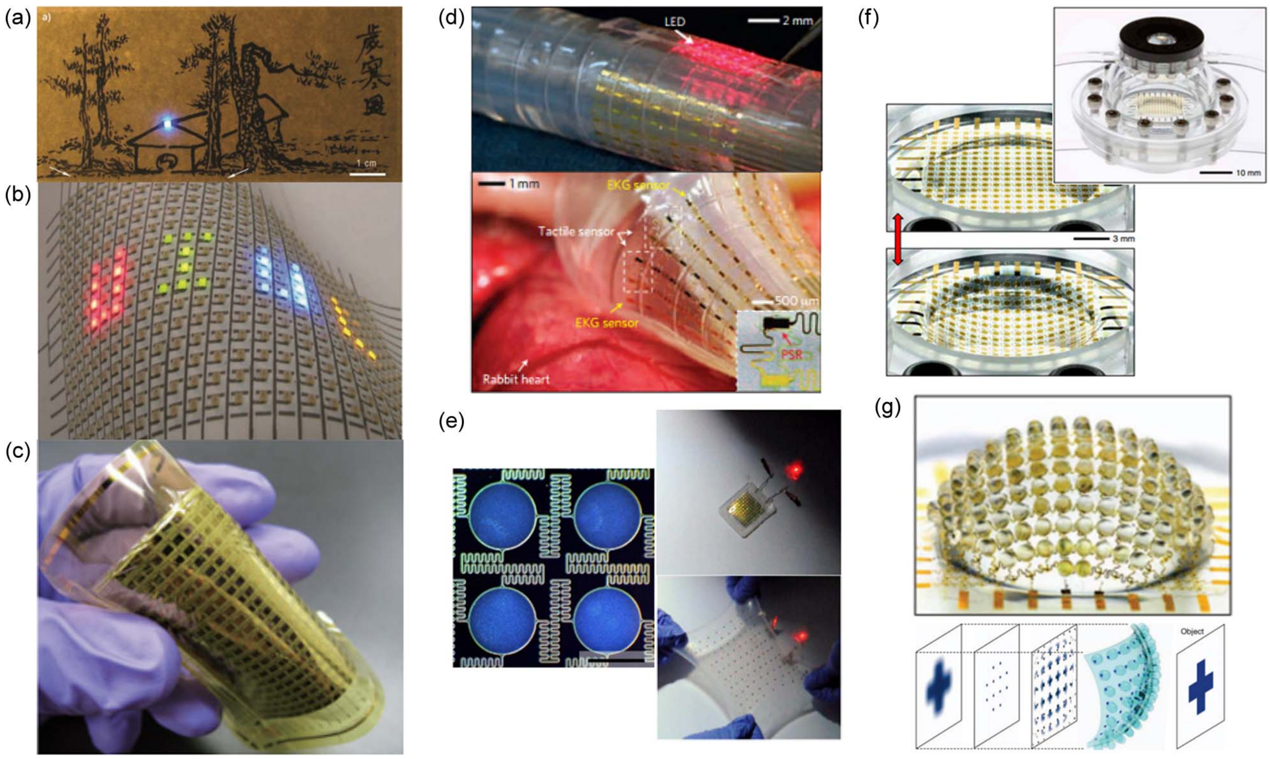

Fig. 4. *(a) Direct-write electronic art, showing a complex trace drawn with a conductive Ag ink on paper, a graphical but functional layout used to successfully power a single LED. Reproduced with permission from [41]. Copyright John Wiley and Sons. (b) A large area printed array of multicolor LEDs as integrated with printed Ag interconnects in a flexible display. Reproduced with permission from [41]. Copyright John Wiley and Sons. (c) Artificial skin comprising Ge/Si core/shell nanowire arrays, here used as channel materials for active-matrix FETs. Reproduced with permission from [98]. Copyright Nature Publishing Group. (d) An instrumented balloon catheter in both its relaxed (upper panel) and inflated states (lower panel). Reproduced with permission from [101]. Copyright Nature Publishing Group. (e) Optical images of the electrode pads and "self-similar" interconnects used for stretchable battery elements, here supported on a Si wafer (left), images showing the battery powering a red LED in relaxed (upper panel, right) and 300% biaxial stretched states (lower panel, right). Reproduced with permission from [102]. Copyright Nature Publishing Group. (f) Pictures of an electronic eye zoom lens camera showing the curvature of sensor array surface being dynamically actuated to match the Petzval surface of plano-convex lens. Reproduced with permission from [4]. Copyright National Academy of Sciences, USA. (g) Image of a model Arthropod eye camera (upper panel) and the schematic description of the mechanisms of image formation (lower panel). Reproduced with permission from [6]. Copyright Nature Publishing Group.*

artificial skin that is capable of sensing and mapping tactile pressure profiles. The devices in this case comprise Ge/Si core/shell nanowire arrays that serve as the channel materials for active-matrix FETs [98]. The fully flexible device spans an area of 7×7 cm^2 including 342 pixels, and can be bent to a radius of 2.5 mm.

Enabling electronic functionalization of flexible/stretchable curvilinear surfaces can deliver unprecedented capabilities of sensing, actuation, and control. For example, balloon catheters are widely used to eliminate stenotic vessel blockage or perform ablation for atrial fibrillation [99], [100]. One limit of the conventional balloon catheters is that little *in-situ* feedback can be provided while an operation is proceeding, and any tendency towards over-inflation may lead to tissue damage. The integration of semiconductor sensor devices with the elastic membrane of the balloon catheter provides a means through which real-time readings of the physiological conditions present in a tissue can be made in benefit to a more effective clinical treatment. An example of an instrumented balloon catheter is shown in Fig. 4(d) [101]. The electronics integrate a variety of sensors in a mesh supported on the elastic membrane, which are capable of measuring temperature, bio-fluid flow, tactile-contacts, and electrophysiological signals. The sensor devices, with active sensors located at the planar nodes of the metal interconnects, tolerate tensile strains up to 200%, while effectively decoupling the sensors from lateral strain upon balloon inflation. Catheters further integrating RF ablation electrodes can excise tissue while monitoring localized heating using the temperature sensors.

An interesting approach to "self-sufficient" electronics has recently been described, an integration strategy that entails the co-integration of power supplies and electronics in ways that match the mechanics and deformations of a flexible substrate material. The images shown in Fig. 4(e) illustrates an interesting example of a stretchable battery-powered device [102]. The layout of the design integrates lithium battery disks (as well as electronics for wireless recharging) that are interconnected by serpentine metal wires—iterating "self-similar" arcs in a mesh network that can undergo large magnitude stretching, folding, and twisting deformations while maintaining high output voltage and capacity, with a record tensile strain up to 300%. This exceptionally compliant power system appears to be especially well suited for use in lightweight and wearable robotics.

An especially exciting application space for flexible/stretchable electronics lies in the area of imaging. Support for this contention can be developed by a consideration of the modes of application of planar imaging media in camera technology, in which optical aberration is usually overcome by adding complexity to the lens system [103], dramatically increasing the weight, bulk and (potentially) cost of the device. For example, a simple lens renders a curved image surface (Petzval surface) that is poorly matched to a planar sensor array, requiring a compound lens system capable of correcting off-axis rays [104]. A curved sensor array that can adapt to the Petzval surface provides an alternative design rule for an aberration-free imaging system [5]. The system shown in Fig. 4(f) illustrates one approach to such a design [4]. The camera (upper panel) integrates a fluidically actuated plano-convex lens whose curvature can be continuously adjusted by the fluid pressure. An array of silicon photo-detectors, transfer-printed and interconnected on a stretchable membrane whose curvature can be similarly varied is mounted beneath it (lower panels of Fig. 4(f)). The assembly forms an adjustable zoom lens that can render an image precisely on the surface of a matching sensor array. The design shown here can be generalized to a model for a camera that embeds a compound lens system reminiscent of those of arthropod eyes [6]. In this case, transfer-printed silicon photodiode arrays supported on an elastic membrane are aligned and bonded to a deformable PDMS micro-lens array. Actuation yields a hemispherical deformation mimicking the compound apposition layouts of arthropod eyes, as illustrated in Fig. 4(g). Precise engineering of the curvature of the system and the configuration of the individual micro-lenses provides a nearly infinite depth of field, together with a wide field of view of about 160° without off axis aberration. Possible applications include ones in intelligence/surveillance and endoscopy. Taken together, these novel designs suggest the potential for important impacts that might be realized based on adoption of advanced materials and manufacturing concepts of flexible/stretchable electronics.

V. BIO-INTEGRATED ELECTRONICS

We finally close with a consideration of the potential impacts that might come from the capability of assembling electronic devices in flexible/stretchable configurations that can innately support their integration with living systems. This represents one of the most challenging scenarios in which any electronic device of this form might be employed. By necessity, they must be compliant enough to laminate on or affix to soft/curvilinear tissues without eliciting deleterious mechanical stresses, yet be sufficiently robust to sustain adhesion, resist moisture and chemical degradation, and withstand fatigue due to tissue motion. Advances have been made to this end, with epidermal electronics [105] representing one of the most explored categories where important medical applications are emerging. Fig. 5(a) shows images of a conformal temperature sensing device that can be directly laminated to human skin for its thermal characterization [106]. The images show a device array—as affixed and under deformation—that consists of arrays of two types of sensors—here 8×8 silicon PIN diodes (higher panel) and 4×4 gold temperature coefficient of resistance (TCR) sensors (lower panel), respectively. The device design adopts an open mesh serpentine layout that effectively releases the strains induced by tensile deformation, and a neutral mechanical plane design in which the active sensors are sandwiched between PI layers, providing an additional strain releasing mechanism upon wrinkling of the skin (images shown in the upper panels). The ultrathin construct renders an effective modulus that matches that of the epidermis. These devices possess a milli-kelvin temperature resolution and can be operated in an imaging mode with a precision comparable to that of infrared cameras, a capability suitable for monitoring the thermogenesis associated with inflammation.

Similar stretchable electronic fabrics can be adapted to provide epidermal sensing and actuating devices for use on organs like the brain [107] and the heart [32], [108] [see Fig. 5(b)]. These arrays can be used to detect and record electrophysiological signals and probe physiochemical states of organ tissues.

A striking use of mechanical compliance in a functional form is illustrated in the images shown in Fig. 5(c). Here the coupling of the energy of stretching motions to the piezoelectric elements of the device provides a way through which mechanical energy can be harvested to supply electrical power to implants, including pacemakers, cardioverter-defibrillators, and heart rate monitors. In the example shown, the expansion and contraction of the heart is captured by arrays of piezoelectric devices, ones comprised of lead zirconium titanate (PZT) micro-ribbons and Schottky bridge rectifiers encapsulated in a biocompatible PI that isolates the electronics from bodily fluids and tissue [109]. Conformal lamination on organs having active contraction and relaxation motions (heart, lung, and diaphragm) yields local strains that activate the PZT devices, with conversion efficiencies (up to 2%) suitable for charging a battery [lower panels of Fig. 5(c)].

Instead of surface-mounted configurations, the delivery of semiconductor devices into the volumetric depths of soft tissue poses another challenging, yet important

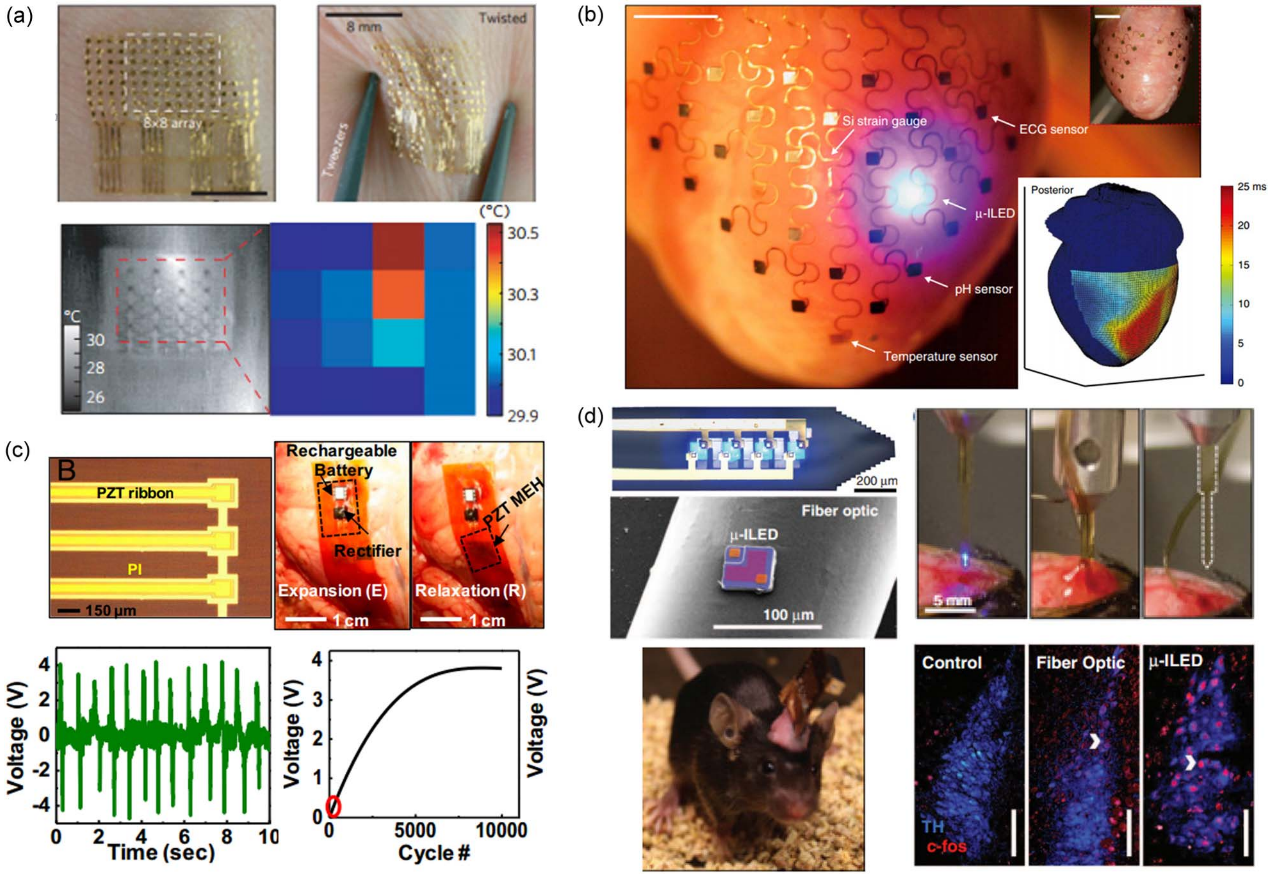

Fig. 5. ***(a) Epidermal temperature sensor array laminated on human skin for thermal characterization (upper panel), with sensitivities comparable to commercial infrared camera and capabilities for imaging-mode measurements (lower panel). Reproduced with permission from [106]. Copyright Nature Publishing Group. (b) Integrated conformal sensor arrays laminated onto a living rabbit's heart. The panel shown at lower right is a spatial map of the electrical activation time measured on the posterior surfaces of the heart. Reproduced with permission from [108]. Copyright Nature Publishing Group. (c) Optical image of piezoelectric PZT ribbons printed onto a PI thin film substrate (upper left) and an integrated device mounted on a bovine heart (upper right), with data showing piezoelectric activation and voltage output coming from piezoelectric energy transformation (lower left); the data given in the panel at the lower right shows battery voltage plotted against time during charging by the PZT system. Reproduced with permission from [109]. Copyright National Academy of Sciences, USA. (d) Integrated electrooptical micro-devices supported on a thin plastic strip; the devices shown have sizes much smaller than 200 μm fiber optic implant more commonly used in optogenetics protocols (upper panel, left). A mouse after device implantation into deep-brain tissue for wireless optogenetics experiments (lower left). The panel at higher right shows the injection of the device into deep structures of the brain. After 1 h of photostimulation, appreciable differences in expressed biomolecular species are found between an implanted fiber optic probe and the flexible integrated device, results suggestive of different levels of trauma/functional response (lower panel, right). Reproduced with permission from [112]. Copyright The American Association for the Advancement of Science.***

opportunity for the use of flexible/stretchable electronics in areas of medicine. Important distinctions can be made between rigid device form factors, such as based on penetrating electrodes [110] or fibers [111], and a soft device that can minimize tissue damage, inflammation, and scarring. These ideas are illustrated in Fig. 5(d), which shows a GaN LED, Si photo-detector, Pt thermo-resistor and a Pt electrode of cellular scale fabricated on narrow, thin plastic strips by established micro-fabrication and transfer-printing techniques (upper panel on left) [112]. They are aligned and stacked in a layer-by-layer manner, resulting in a flexible, multifunctional device of a total thickness of $\sim$20 μm. Owing to its compliant nature, the ultrathin device is inserted into deep tissue using a releasable injection needle in a minimally invasive operation, and once placed can deliver photons to a targeted location with high spatial and orientational precision. The data coming from these studies, which reveal significant differences in biological response as compared to fiber optic probes, suggest that devices of this type will provide a powerful new tool for use in the field of optogenetics.

VI. CONCLUSION

As discussed in this review, the emerging design strategies, advanced materials in the form of high performance inorganic EM inks, and enabling means of fabrication via printing and transfer assembly are providing a practical foundation for the development of flexible/stretchable electronics with far ranging potential for engendering impacts in technology. The devices so enabled provide useful, new, and in many instances previously difficult to realize functional capabilities for electronics while retaining levels of performance more typically associated with chip-based, high-integration electronics. It is now well appreciated that the progress being made in research on unconventional form factors for electronics will likely transform many areas of technology, with perhaps the most profound impacts being felt in areas of consumer electronics—flexible light weight displays, wearable electronics being notable examples. The work highlighted above supports this assessment, but also suggests an importance for technologies that will be quite different in terms of the nature of the manufacturing regime they would need for commercial success. In our view, the development of an enabling electronics technology that seamlessly merges the form factor of high performance electronics with those of complex systems and phenomena—the soft, adaptive, and hierarchical forms of living systems or the curvilinear designs that innately mesh with optics, as examples—stands as both a frontier challenge for engineering research and significant opportunity for progress in technology. As the highlights given above suggest, the progress made in a very short time is remarkable. More importantly, though, the pace of advancement towards commercialized objectives is clearly accelerating. All the same, we have only begun to imagine what might be possible. ■

Acknowledgment

This work was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, under Award # DE-FG02-07ER46471.

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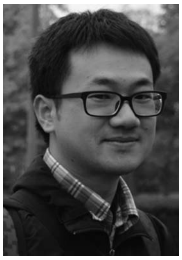

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