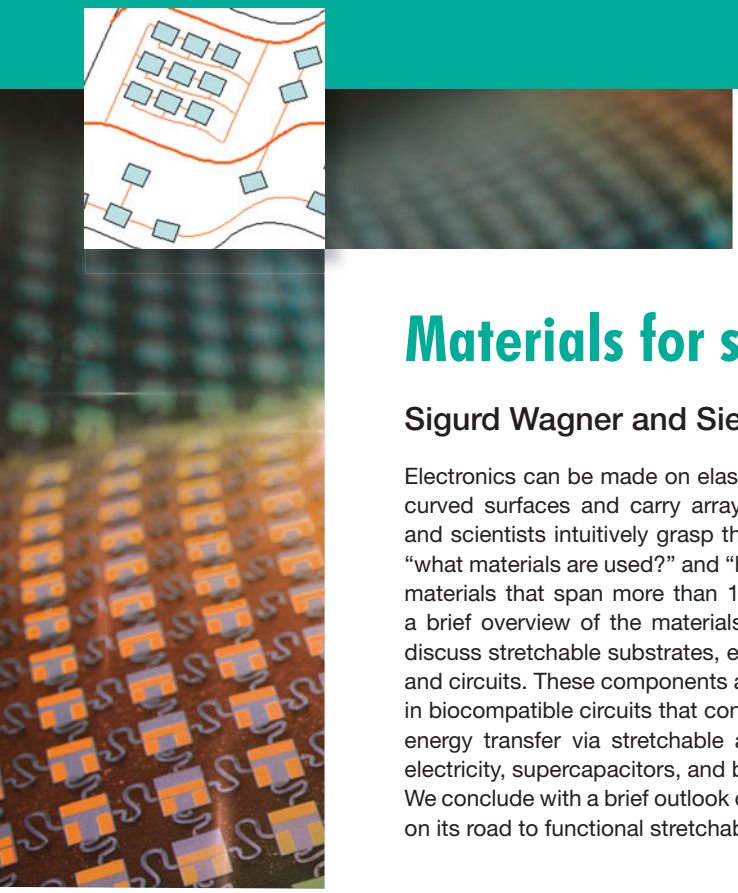

Materials for stretchable electronics

Sigurd Wagner and Siegfried Bauer, Guest Editors

Electronics can be made on elastically stretchable "skin." Such skins conform to irregularly curved surfaces and carry arrays of thin-film devices and integrated circuits. Laypeople and scientists intuitively grasp the concept of electronic skins; material scientists then ask "what materials are used?" and "how does it work?" Stretchable circuits are made of diverse materials that span more than 12 orders of magnitude in elastic modulus. We begin with a brief overview of the materials and the architecture of stretchable electronics, then we discuss stretchable substrates, encapsulation, interconnects, and the fabrication of devices and circuits. These components and techniques provide the tools for creating new concepts in biocompatible circuits that conform to and stretch with living tissue. They enable wireless energy transfer via stretchable antennas, stretchable solar cells that convert sunlight to electricity, supercapacitors, and batteries that store energy in stretchable electronic devices. We conclude with a brief outlook on the technical challenges for this revolutionary technology on its road to functional stretchable electronic systems.

Stretchable electronics, the newest class of large-area electronics

Stretchable electronics is the newest class of large-area electronics. Quite literally large-area electronics has become a success story: flat panel displays are manufactured at the rate of 100 and solar cells at 200 square kilometers a year. Many of these products are made with thin films. Liquid-crystal displays[1] are driven by active matrices of amorphous silicon transistors,[2] and 10% of all solar cells are made of amorphous silicon[3] or chalcogenide films.[4,5] Like any other stiff material, circuits become flexible and rollable when their thickness is reduced to 1/1000 of the desired radius of curvature.[6–9] Thin-film circuits are made on flat surfaces by standard microfabrication techniques. When made on plastic substrates and with plastically deformable interconnects, they can be shaped to surfaces that need expansion out of the plane, for instance spherical caps.[10] On plastic substrates, this deformation is permanent. Elastomeric substrates and elastic interconnects let circuits go a shape beyond: to reversible deformation and near-arbitrary dimensions.[11–16] Sizes and shapes of elastomeric circuits can be changed reversibly by applying mechanical force,[17] by gas pressure,[18,19] or by application of an electric field.[20,21]

Now we can make electronic skin,[22] conformable sensors and displays (see the Kim et al. and Sekitani and Someya articles in this issue), electronic biointerfaces,[12,23–25] electronic muscles,[20,21,26] and energy harvesters (see the article by Kornbluh et al. in this issue). Bending,[7–9] shaping,[10,27] stretching (see the Suo article),[11] and electroactuation[20,21] or energy harvesting are illustrated in **Figure 1**. The latest progression is circuits made on a biodegradable substrate,[28,29–31] which disappears to leave the bare circuit conforming to a living organism; the goal is a circuit that envelopes the heart fully to sense and stimulate it with high spatial resolution while expanding and shrinking with the heartbeat.[31] Stretchable electronics is the research frontier of large-area electronics.

A materials scientist first will notice the elastic, soft nature of experimental stretchable electronics: a polymeric electro-actuator (see the article by Kornbluh et al. in this issue), a wraparound pressure sensing skin (see the article by Sekitani and Someya), or a neural interface in the folds of the cerebral cortex (see the article by Kim et al.). The materials scientist will then ask: How does this work? How can I make this? Stretchable electronics are fabricated with planar technologies that evolved from the materials and processes of classical microfabrication to those of large-area electronics on rigid substrates, and then they are branched out to a diverse range of fabrication techniques. Functionalities never seen before can be realized by combining electrical with mechanical properties drawn from all conceivable classes of materials: liquid, gel, solid, organic and inorganic, insulator, semiconductor, and metal.

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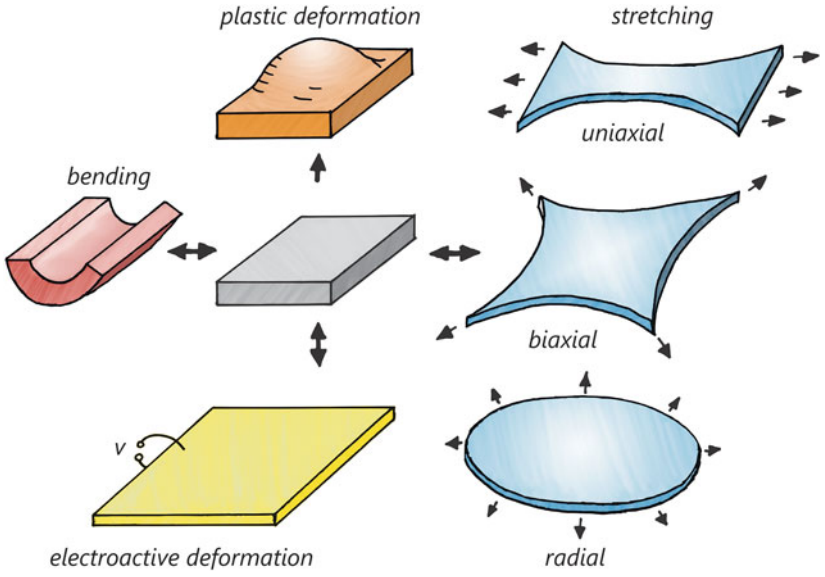

Figure 1. A substrate may be permanently shaped, bent, or stretched uniaxially, biaxially, or radially by mechanical force. Charging a capacitor with an elastomeric dielectric to a voltage *V* compresses the dielectric, which converts electrical into mechanical energy. Used inversely, the capacitor works as a charge pump that generates electrical energy.

A diversity of materials

The Young's modulus *E* of the materials for stretchable electronics spans a range of at least 12 orders of magnitude. In general, both internal energy and entropy change upon mechanical deformation of materials. Materials used in stretchable electronics are comprised of extremes, from entropy elastic elastomers to energy elastic solids (see the article by Suo in this issue). Stretchable electronics combine liquids, soft and elastic materials, and hard and brittle materials. In **Figure 2**, the Young's modulus provides an instructive scale for our survey of materials. Gels compare to brain tissue (200 Pa) and muscles (1000–3000 Pa); the typical polydimethylsiloxane (PDMS) elastomer, electroactive elastomers, and other rubbery materials range from 1 to 100 MPa; and polymers and organic semiconductors have moduli in the GPa range. Elastomers with a low Young's modulus *E* in the range of 1–100 MPa have a bulk modulus *K* on the order of 1–10 GPa, which is comparable to those of typical polymers. Such highly elastic materials are easily stretched by mechanical forces, while their volume stays constant. In contrast, solids typically have high Young's moduli, and they cannot be stretched reversibly by much, or else they will deform plastically or break. Gold (70 GPa), silicon (150 GPa), one of the hardest solids, diamond-like carbon (DLC, 500 GPa), and one of the strongest materials, graphene (1000 GPa along its plane), complete the spectrum of materials. The regimes of elastic, plastic, and brittle materials also are evident in Figure 2. Combining elastic substrates and interconnects with plastic or brittle device materials[32,33] sets up tremendous mechanical contrasts, with amazing consequences. A hard solid, when bonded to a polymer, can become surprisingly deformable. Suo explains how such mechanical contrasts have inspired mechanics theory (see the article by Suo).

The architecture of stretchable electronics

Stretchable electronics are designed for uniaxial, biaxial, or radial stretching by tens of percent. But optoelectronic devices and circuits contain brittle materials that will break if stretched by more than a few tenths of a percent. For their protection, the circuits are placed on platforms, or islands. Most islands are rigid in-plane and are flexible out-of-plane because they are thinner than ~1/1000 of the typical bending radius (e.g., 1 μm versus 1 mm), as illustrated in **Figure 3**.[32] The substrates exposed between the rigid islands, together with the conductors that run across it, accommodate the elastic strain. The islands are made of one of the circuit materials (see the Kim et al. article),[33] of separate thin-film materials,[34] or of stiff plastic foil (see the Sekitani and Someya article).[13] In a macroscopic approach to building stretchable systems, conventional rigid printed circuit board "islands" are connected with stretchable

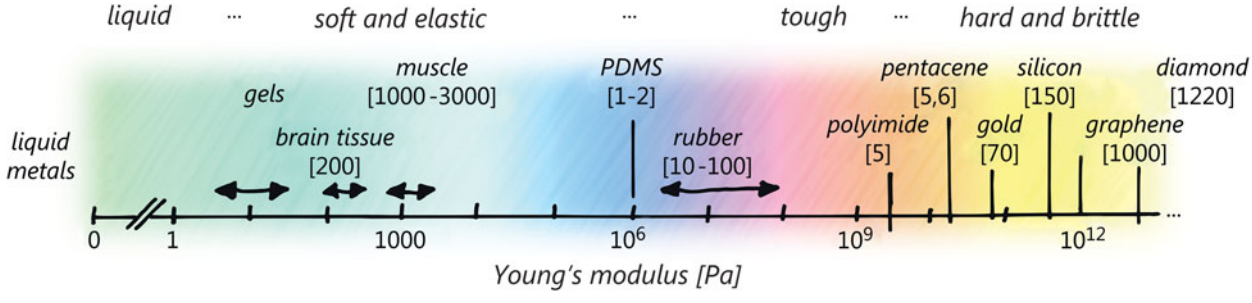

Figure 2. Stretchable electronic surfaces combine viscoelastic, plastic, and brittle materials with many orders of magnitude differences in Young's modulus. The materials range from liquid metals, gels, brain tissue, muscles, elastomers, polymers, metals, semiconductors, to carbon-based solids. Note: PDMS, polydimethylsiloxane. The numbers in brackets are in Pa for soft, MPa for tough, and GPa for hard materials, respectively.

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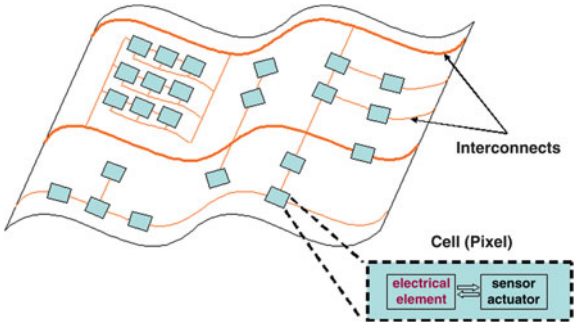

Figure 3. Architecture of an elastic electronic surface. Cells with optoelectronic functions are made on islands and are connected with flexible conductors on a flexible substrate.[32]

copper conductors[35] (see the article by Vanfleteren et al. in this issue) in a design similar to that of rigid-flex circuits, where circuit boards are connected by flat cables.[36] When the whole system is stretched, the substrate, devices, interconnects, and protective encapsulating layers interact. In the following, we separate these components for the purpose of introducing them one by one.

Stretchable substrates and encapsulation

We can make substrates stretchable in three ways, as illustrated in **Figure 4**: (1) by stretching an elastomeric membrane, (2) through a net made of an elastomer or of a polymer, or (3) by stretching the polymer foil of a flex circuit flat. Polydimethylsiloxane (PDMS) is the elastomer of choice because it can be made with widely accessible and available laboratory techniques, is also commercially available, and only swells in but is not attacked by most process chemicals[37] (although it is etched by KOH and HF). Acrylics also are used as passive substrates similar to PDMS[38] and as electroactive materials (also see the Kornbluh et al. article).[19–21,39] Nets are punched out of polyimide foil[25,40] or molded from PDMS.[41] The well-established flex circuits are the equivalent of printed wiring boards but made on plastic foil.[36] The entire flex circuit can be folded and unfolded. Such flex circuits need only bendable[42,43] instead of elastically stretchable encapsulation. For stretchable organic electronics, some of which are sensitive to atmospheric corrosion, that is an important advantage because no highly hermetic *and* stretchable permeation barrier has been invented yet.

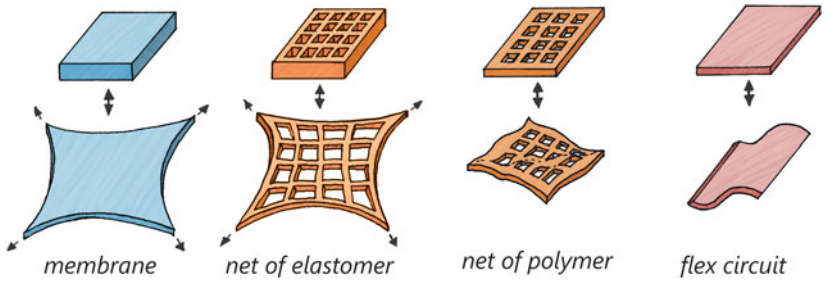

Figure 4. Substrates for skin-like stretchable electronics are either elastomeric membranes or nets made of elastomer or polymer foil. Flex circuits are made on polymer foil.

In stretchable electronics, devices are subject to alternating tensile and compressive strain. This is illustrated with the following example of a membrane that is stretched uniaxially and then relaxed again. Extending a long membrane stripe of length L by pulling out its opposite ends to a length $L\,(1+\varepsilon)$, where ε is the strain, narrows the stripe from width W to $W(1-\varepsilon/2)$, and thins it from thickness H to $H(1-\varepsilon/2)$. Therefore, the device islands and interconnects on the membrane experience lateral compression. When the substrate is relaxed, it experiences lateral tension. The surface area of the substrate increases to $LW(1+\varepsilon/2)$ during uniaxial stretching to strain ε. Biaxial or radial stretching[44–46] to a linear strain of ε raises the surface area to $\sim LW(1+2\varepsilon)$. Hence, for a given value of linear strain ε, biaxially or radially stretching the substrate puts the device islands and interconnects under four times the stress that is developed in uniaxial stretching. In experiments, membranes can be stretched uniaxially to $\varepsilon = 0.5$ and more without breaking devices or interconnects, while going above linear $\varepsilon \cong 0.15$ in biaxial or radial stretching is a challenge.

For mechanical and chemical protection, stretchable electronics are encapsulated with the same or a similar material and with the same processes, as employed for the substrate. The encapsulant is dispensed as a liquid and is then cured or laminated on. Stretchable biointerfaces on PDMS substrates are encapsulated with photopatternable silicone or with PDMS.[22,47,48] Freestanding stretchable conductors are first encased in polyimide and then in PDMS for better mechanical matching with the metal conductor (see the Kim et al. and Vanfleteren et al. articles). Contact holes through the encapsulation are opened by plasma etching,[47] liftoff,[49] or lithography of a photopatternable elastomer.[50,51] Contacts at the edges of the stretchable membrane are opened with procedures similar to liftoff.

Stretchable interconnects

Stretchable interconnects combine stretchability with high electrical conductance. While thin-film or thinned integrated circuits may need thin-film interconnects, hybrid circuits that include conventional devices can make use of relatively thick printed polymer filled with conducting particles or of copper laminate sheet. Stretchable conductors rely on configurations that are illustrated in **Figure 5**: waves;[11,13,39,53] meanders;[14,35] helices of metal on/in/around a stretchable substrate;[54] spirals of silicon;[55] percolation through conducting particles embedded in a stretchable matrix or substrate;[25,52] nets with meshes on the micrometer,[56–58] multi-micrometer,[31] or millimeter[59] scales; and liquid metal confined within a microfluidic circuit made of PDMS.[60] These conductors fall into two groups of stretchability up to electrical failure. When placed on or embedded in an elastomer such as PDMS or a gel, some conductors inherently are stretchable to large strains. This is the case with ion-implanted PDMS and carbon nanotube-filled

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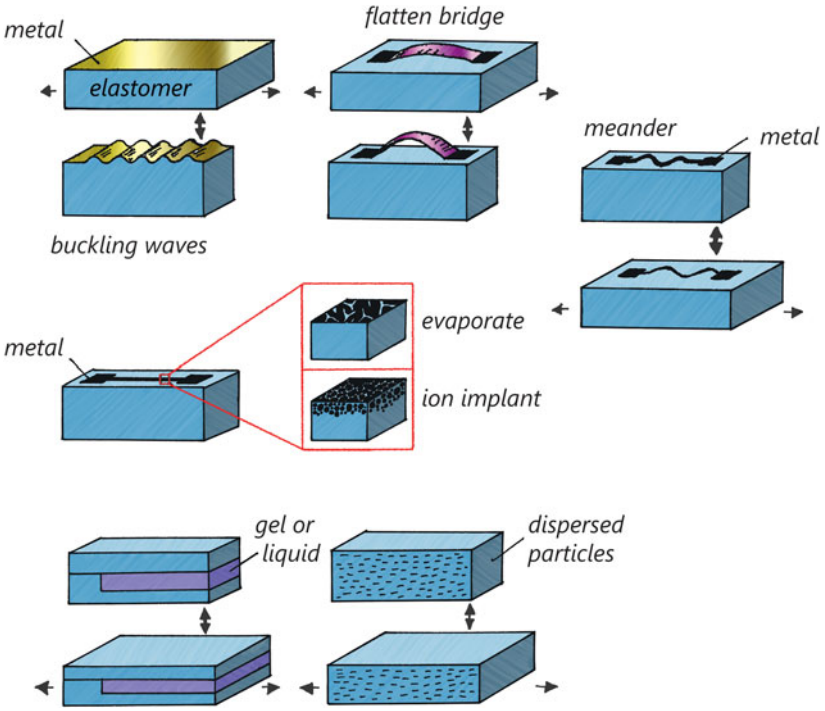

Figure 5. Techniques for making stretchable interconnects; clockwise from upper left. Waves or bridges of a conductor; meanders (or spirals); conductor filled polymer or gel, or microfluidic metal; and percolation through the network of a surface film or of a shallow implant.

gel, as well as stretchable gold nets on the micrometer or millimeter scales. Such inherently stretchable conductors need no special preparation for uniaxial or biaxial stretching. A second group of conductors relies on waves that form after a stiff material—a surface coating, conductor, device, or circuit—has been applied to the pre-stretched elastomer. Upon relaxation from pre-stretch, the stiff material buckles to waves as it experiences compression by the contracting substrate. Stretching and relaxing this substrate uniaxially, in the pre-stretch direction, reversibly stretches and relaxes the surface wave.

The shear force at the interface between substrate and device material must not exceed the strength of adhesion, or else the device will peel off when it is compressed during relaxation. When the shear force reaches through the thickness of the substrate, the whole structure will coil up. The highly perfect surface waves that can be made on PDMS[61,62] lend themselves to tunable diffraction gratings[63] and wavelength-tunable optically pumped lasers.[64]

Stretching pre-formed waves,[11,31,65] meanders,[14,35] and spirals[54] develops smaller strains in the conductor than stretching inherently stretchable materials.[66] When a complete flexible circuit net of silicon islands and interconnects is transfer printed on a chemically patterned, pre-stretched substrate, the silicon links that are coated with interconnect metal arch up to bridges.[31] As the substrate is stretched, these connecting bridges are pulled down toward the substrate. Such up-and-down moving bridges avoid the Poisson compression and expansion of interconnects that are integrated with the elastomer, which may open cracks along the stretching direction of interconnects. One advantage of the surface-integrated conductors is their ease of encapsulation. Preventing the substrate from narrowing during pre-stretch produces fully integrated conductors that are free of cracks.[62] Uniaxial pre-stretching makes the conductor uniaxially stretchable, and biaxial or radial pre-stretching or heating renders it biaxially stretchable.[44,46,61,67] Meanders and spirals can be stretched in only one direction. The deformation of nets on both the micrometer and millimeter scales resembles that of linked meanders,[56,66] with the links enabling stretching in any direction. Meshes of metallic nets on the micrometer scale have been observed to lengthen and shrink during their stretching and relaxation.[52]

The two important performance parameters for stretchable interconnects are high electrical conductance and large critical strain at which conduction is lost. During stretching, the electrical connection is interrupted when particles disconnect from each other, waves, meanders, spirals, or nets break, or liquid escapes. The critical strain depends on conductor technology and whether the stretching is uniaxial or biaxial. Mechanical prestretching produces structures than can be stretched up to ~100% uniaxially and up to ~15% biaxially. For electrodes on electroactive polymers, a low Young's modulus, or more precisely, the elastic modulus in the case of a nonlinear elastomer, becomes an equally important third parameter: during their cycles between electrical and mechanical energy, the electrodes must store as little elastic energy as possible, as this energy is robbed from useful conversion. Corrugated metal electrodes meet this requirement (see the article by Kornbluh et al.).

Devices and circuits

Broadly speaking, components, devices, and circuits are placed on stretchable substrates by integration or by lamination, as illustrated in **Figure 6**. For direct integration with the substrate, component materials are deposited and patterned layer-by-layer on the elastomer. This process is similar to the fabrication of conventional wafer-based or thin-film integrated circuits. Patterns are formed either subtractively, that is deposition followed by selective removal, or additively, by the direct printing of patterns. In lamination, components or complete circuits are adhered to the elastomer with adhesives or by transfer printing. The components are pre-fabricated elsewhere on flexible plastic substrates, thin flexible wafers, or rigid islands.

Direct deposition and subtractive processing at present are seen as the pathway to wallpaper-size stretchable systems[68,69]

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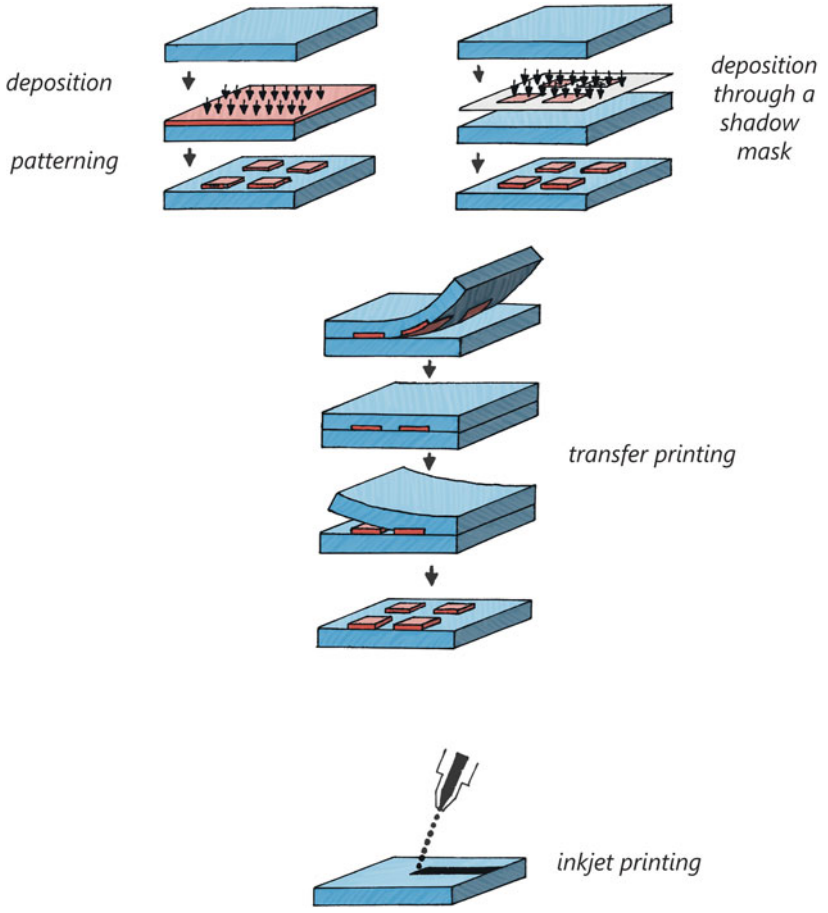

Figure 6. Techniques for fabricating devices on elastomeric substrates: deposition followed by lithographic patterning, deposition through a patterned shadow mask, transfer of devices by printing, and additive printing.

because of their success in flat-panel display fabrication. High-speed additive printing of optoelectronic circuits eventually may displace these batch fabrication processes. As a substrate, PDMS is quite different from wafer silicon or plate glass. Photolithography and electronic inks employ solvents that make PDMS swell, which in turn may delaminate device films. Converting these processes to alternative solvents takes time.[48] Given the high coefficient of thermal expansion of PDMS, ~300 ppm/°C, its dimensions may vary noticeably during the temperature cycles associated with processing. Lamination, which has the advantage of minimal on-elastomer processing, produces hybrid circuits that are similar to conventional printed wiring boards and flex circuits. The comparative ease of lamination and hybrid assembly has enabled impressive demonstrations of quite complex stretchable optoelectronic system concepts (see the Kim et al. and Sekitani and Someya articles). Eventually, stretchable systems will combine large integrated arrays of simple devices with nodes of nanoscale integrated circuits in complementary metal-oxide-semiconductor (CMOS) technology to provide large-area, high-resolution, high-performance sensor skins. In this architecture, human-size tasks, such as high-resolution multipoint sensing, will be assumed by electronics directly integrated on the substrate, and tasks that demand high power and high speed computation and communication will be executed by laminated CMOS circuits. Lamination of another kind is sought for biological applications,[12,23,27,44] where the adhesion of healthy cells or of a tissue culture to the stretchable device is a basic measure of the quality of the electronics/biointerface.

Stretching substrates with rigid circuit islands sets up non-uniform mechanical strain: small strain on and immediately beneath the island, larger strain on the surface between the islands than at the bottom of the elastomer, and largest strain in the elastomer at the periphery of the island.[70,71] Figure 3a of Suo's article in this issue shows one consequence of the high strain at the edge of a silicon nitride island patterned on a polyimide foil. The edge of the island slipped back when the foil was expanded. This slip can occur only if the periphery of the island simultaneously debonds from the substrate.[71] At such transitions between soft and hard materials, surface-integrated interconnects experience their biggest strain and must be designed and fabricated to accommodate it. This situation has been analyzed for plastically deformable substrates (see the Suo article) and is under study for elastomers. Excessive strain is distributed across soft/hard interfaces by grading mechanical strength.[72] The edges of device islands are feathered out,[68] the elastomer below the island is hardened chemically[33] or by insertion of rigid plates,[33] or the interconnect is made to vary from wide and stiff on the island to narrow and compliant on the elastomer (see the Vanfleteren et al. article).

Research on stretchable systems has been dominated by the study of fundamental mechanics, development of fabrication technologies, and demonstrations of the functions they enable. For industrialization, a full toolset of process sheets, models, and simulation techniques also will be needed. Excellent initial steps have been taken in mechanical theory and simulation. Many other steps that lead from material to system, including the architecture of electronic surfaces of variable topography, are waiting for exploration by engaged researchers.

In this issue

Because the research directions of this issue's contributors complement each other, their articles provide a comprehensive overview of the field of stretchable electronics. Suo analyzes the mechanics of stretchable substrates, of electroactive polymers, and of hydrogels, and he announces the era of soft machines. Kim et al. unite imaginative geometries for stretchable interconnects with the high performance of wafer-based electronics and bring

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them to bioinspired and biointegrated applications. Sekitani and Someya craft stretchable nets and polymer conductors for conformable sensor and organic light-emitting arrays, which operate with organic circuits designed to match wafer-based silicon electronics. Kornbluh et al. describe the principles, materials, and configurations of electric power generators, including the polymer engine generator, made of dielectric elastomers. Vanfleteren et al. explain the design, fabrication, testing, and application of stretchable copper meander conductors for interconnecting printed circuit boards, made in a configuration geared to immediate industrial application.

Outlook

Each researcher in the field of stretchable electronics has experienced the immediate connect with audiences to whom they present and demonstrate their work. Everyone, from laypeople to experts, intuitively grasps the concept of soft, conformable systems. Researchers are attracted by this instinctive appeal too. But what drives them is the vision of applications that lie beyond the reach of today's optoelectronic technologies. This vision is documented by the contributors to this issue, who created many of the concepts, techniques, devices, and demonstrations of stretchable electronics.

The science that lies along the paths to applications starts with materials and their mechanics, continues to processes, optoelectronics, sensors and actuators, and enables the technologies for making new systems architectures. Stretchable electronics is qualitatively new territory for electronics. The field is developing new concepts for energy transfer, energy supply and distribution, and generation and storage. It is coming up with innovations in wireless energy transfer via stretchable antennas,[73] stretchable solar cells that convert sunlight to electricity,[74–76] supercapacitors,[77,78] and batteries[38] that store energy in stretchable electronic devices. We hope that readers will enjoy this issue and take away stimulating ideas for their own research.

Acknowledgments

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