Needs and Enabling Technologies for Stretchable Electronics Commercialization

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<u>Abstract</u>

Stretchable electronics represent an emerging class of devices that can be compressed, twisted and conform to very complicated shapes. The mechanical and electrical compliances of the technology promise to open up applications for healthcare, energy and entertainment purposes. However, advancement in the field has been hindered by material related constraints. Moreover, the current microfabrication facilities are optimized for rigid substrates such as silicon, which have significant different properties compared to elastomers. In this paper, four categories of enabling technologies for stretchable electronics commercialization are critically reviewed, namely: the novel design of stretchable structures, use of non-conventional materials, state-of-art printing techniques and also the patterning of electrodes or metal interconnects via conventional manufacturing techniques.

Introduction

Multidisciplinary research has paved ways towards a new form of electronics which is not only flexible but also conformable and deformable; while ideally maintaining excellent electrical properties under strain. Known as 'stretchable electronics', new devices such as sensors, transistors and circuits, lighting elements, power sources, as well as their interconnections, exhibit closer resemblance to the human body than their rigid counterparts in terms of mechanical properties^{1,2,3}. Thus, stretchable electronics can improve not only existing technologies but also enable a wide range of novel applications. For example, thin micro-electrodes made on soft and elastic substrate have been developed for neural prosthetics, neural signal recording and neuromodulation^{3,4}. Compared to their rigid counterparts, these neural electrodes show improved mechanical compliance, offered better structural interface with the nervous system and are highly robust. Artificial skin have also been fabricated to detect external stimuli such as pressure, temperature, strain and humidity, which have huge implications for robotics and also amputated patients⁵. Other devices such as epidermal sensors for vital sign monitoring and stretchable energy harvesting for selfpowered electronics have also been shown^{6,7}. More recently, stretchable electronics have found their ways into the entertainment realm. Kim et al. had made a highly stretchable touchpad that can be worn around the arm for new gaming experiences⁸.

Stretchable electronics are rapidly growing at a compound annual growth rate (CAGR) of 121.3%, forecasted to reach \$411.85m in 2023⁹. However, when considering mechanically compliant smart patches and implantable electronic devices for both biosensing and drug delivery applications in personalized theranostic and stratified medicine platforms, market data are less clear. According to Future Markets, potential addressable market for the introduction of these technologies into commercial streams or emerging markets form an opportunity worth between \$8.2bn and \$16.2bn, when penetration rates varying between 20% and 50% are considered, in: *i*) disposable medical sensor, *ii*) blood glucose monitoring, *iii*)

cosmetic patches and *iv*) medical smart patches whose market introduction is expected by 2020^{10} .

The technological hurdles that hinder the commercialization of stretchable electronics include the use of elastomeric materials and the fabrication processes of these devices. Considerable progress has been made to advance this field. In this paper, we cover some of the innovations that enable the development of these devices, which include the novel design of stretchable structures, use of non-conventional materials, state-of-art printing techniques and also the patterning of electrodes or metal interconnects via conventional manufacturing techniques.

Stretchable Structures: Devices and Interconnects

Since early studies on the mechanical compliance of human skin, it has been clear that electronics integrated on the human skin must undergo strains of up to 30% without constraining natural motions¹¹. Reaching this level of stretchability or higher also ensures that the elastic behaviour of human skin can be mimicked, so that a wearable device in form of electronic tattoo or smart patch can perform reliably when the substrate is subjected to the same level of strain. Moreover, implantable electronic devices designed to interact directly with organs, such as the heart muscle, the spinal cord or the brain, are often required to show the same mechanical compliance as the organ itself. This means that actual electronic interfaces need to be fabricated with biocompatible materials having the same values of Young modulus of the target organ. This in practice justifies the scientific and industrial interests in looking at soft electronic devices that can be stretched up to 100% or more of their initial dimensions within their elastic range, so that any changes in terms of their physical (i.e. mechanical, electrical, optical) characteristics are reversible under multiple cycles. Finally, sensors and electrodes on compliant or inflatable balloon catheters for minimally invasive surgical procedures require even higher levels of deformation, i.e. strains of well over $100\%^{12}$.

Stretchability can be achieved with conventional hard, rigid or brittle materials used in the electronic industry via novel device architectures or bespoke design of their interconnecting structures. In an early demonstration of the concept, Khang *et al.* had fabricated stretchable Si MOSFETs and p-n diodes on thin silicon ribbons bonded to polydimethylsiloxane (PDMS)¹³. The multistep process first started with the patterning of silicon ribbons on a Si-on-insulator (SOI) wafer. These structures with only their two ends connected to the wafer were subsequently brought into contact with a pre-stretched PDMS. Upon release of the strain, the silicon formed into wavy structures with defined wavelength and amplitudes that accommodates deformation¹⁴. In the study, devices maintained their properties even after 100 cycles of compression and stretching. Recent examples that utilize the same post-buckling strategy to create wavy structures include highly stretchable arrays of nanostructures and sheath core fibers for superelastic electronics, sensors and muscles^{15,16}.

However, as opposite to non-ceramic based materials that are commonly adopted in printed and flexible electronics, crystalline silicon, thinned down to just few microns in order to become flexible, shows several piezoeffects, both on passive and active devices. These include piezoresistivity in silicon-based resistors, piezojunction effects in bipolar transistors, current sensitivity to magnetic field (PiezoHall), piezotunneling in tunnelling diodes, and change in carrier mobility of MOSFET devices (piezoMOS) under bending¹⁷.

Another approach is to obtain stretchability by distributing active devices on rigid subcircuit islands over an elastomer as shown in figure 1¹⁸. Kim *et al.* had further built on this method by introducing out-of-plane arc shaped interconnects for islands of active devices¹⁹. This can be done by attaching only the device islands onto a pre-stretched PDMS with prior

surface activation of the elastomer via oxygen plasma. Once the stress was released, the compressive forces caused the unattached interconnects to protrude and thus, forming arc-shape structures which can be deformed. By introducing serpentine shapes, it was further shown by the authors that the electronic systems built on the PDMS can be strained up to 100% reversibly through this technique. When these structures are stretched within their limit, the electrical properties of the system approach its planar counterparts as the serpentine interconnects compensate the applied strained (stable up to 1,000 cycles) through the change in height and geometry.

Other structures that accommodate stretching include planar horseshoe interconnects²⁰. Important design parameters include width of the track, radius of curvature and turning angle of the horseshoe tracks as shown in figure 2. More recently, Fan *et al.* had explored fractal design concepts such as Peano, Greek cross and Vicsek for stretchable electronics²¹. The advantage of these layouts includes the endurance to withstand biaxial, radial and other deformation mode.



Figure 1. Stretchable device architecture on a polymer substrate (a) Active devices are fabricated on rigid subcircuit islands connected via stretchable interconnects (b) Cross-section of the electronic surface with strain profiles when stretched. Reproduced with permission from Ref ¹⁸ Copyright 2005 IEEE.



Figure 2. Stretchable horseshoe interconnect (a) geometrical parameters where r = radius of curvature, $\theta = turning$ degrees, W = track width, $W_o = overall$ width. (b) horseshoe interconnect of different turning degrees. Reproduced with permission from Ref²⁰ Copyright 2012 Cambridge University Press.

Materials for Stretchable Electronics

Due to the intrinsic low strain capability of conventional materials, alternative materials have been explored. Polymeric materials such as PDMS and polyurethane are commonly used as alternative substrates to silicon wafers²². While the majority of organic materials can generally be rendered flexible, such materials are not stretchable, which is a key mechanical property necessary to realize applications of E-skin for prosthetics, artificial intelligence, systems for robotics, personal health monitoring, biocompatibility, and communication devices. In order to overcome these limitations, intrinsically stretchable and self-healing polymeric materials for electronics, including conducting, dielectric and semiconducting polymer have been reported^{23,24}. Researchers have also developed a wide range of conductors that are intrinsically deformable, including metal based, carbon nanocomposite based and conducting polymer based materials.

In the case of metal-based inks, for instance, film conductivity is given by their formulation, containing conductive material in form of micro or nanoparticles, a binder and solvent, so that conductive particles are bonded together to form a conductive film when the solvent leaves the ink after deposition, which rebuild the bonding at molecular and atomic levels^{25, 26}. Therefore, if the electrical performances of printed inks are nowadays approaching those of the same materials deposited with conventional methods, the electro-mechanical

properties of materials deposited by printing might be substantially different. However, not all inks require the presence of binder and solvent. One such example the liquid metal alloy of gallium, indium and tin, known as 'Galinstan', that can be used directly as a conductive ink without requiring any solvent thanks to its low melting temperature at the eutectic composition²⁷. The fluidity may remain after the ink has been printed, which enhances the stretchability, but this does however require further packaging²⁸.

Other promising composites have been reported with carbon-based inks. Gao *et al.* had printed pristine graphene ink which were still conductive after 1000 bending cycles under a folding angle of $180^{\circ 29}$. Highly conductive elastic conductors have been reported by Chun *et al.*³⁰. The metal and carbon nanotubes hybrid composites displayed a conductivity of 5710 S cm⁻¹ and can be stretched up to 140% strain. Sekiguchi *et al.* had fabricated elastomeric field effect transistors based on single-walled carbon nanotubes which could be stretched (elasticity: ~ 110%), bent, compressed (>4.0 MPa, by a car and heels), impacted (>6.26 kg m/s, by a hammer), and laundered³¹. Many of the inks need post-deposition treatment, such as sintering, in order to maximize their functionality²⁵.

Printing Technologies for Stretchable Electronics

Printing technologies represent some of the enabling tools for the adoption of the aforementioned inks. Compared to conventional microfabrication processes, printing technologies are potentially cheaper and are able to deposit a wide range of both active and passive materials onto a generic planar or conformable substrate³². Three printing approaches: screen printing, inkjet printing and, more recently, aerosol jet printing is discussed here.

Screen printing is a well-established approach that utilizes a screen of woven material patterned with different sizes of the mesh and positioned at short distance from the substrate to be printed on. When ink is spread over the screen by squeegee, the ink is transferred from the screen to the substrate underneath according to the pattern contained in the screen³³. One of the characteristics of screen printing process is related to direct contact of screen and substrate during printing, whose limitations include control of accuracy, especially on soft substrates, and potential contamination on bio-compatible substrate applications.

On the other hand, jet printing methods, also called "direct writing" provide non-contact printing condition that can minimize the stated issues. Thickness and resolution are two important aspects for high performance stretchable electrodes. It has been decades since the first inkjet printing technique was commercialised back in the seventies, and the technique has been diversely developed and well-established for precisely delivering tiny amount of liquid inks at relatively high resolution with accurate automation control. A common structure of the technique includes an inkjet printhead that has a small nozzle aperture on the tip, with a typical nozzle size of 10-50 μ m³⁴. Ink is driven through the printhead from where it forms droplets (20-100 μ m) for deposition on the substrate. The process can be further assisted by heating, airflow, electric field or acoustic waves, to minimize the size of droplet formation³⁵. The simple structure of the inkjet printhead allows researchers to fabricate proprietary printheads to meet special needs²⁸. A wide range of materials can be formulated for inkjet printing, including metallic nanoparticle inks, carbon inks, polymers or inorganic particle inks. However, due to the direct contact between the ink and the nozzle, the tolerance of ink viscosity is often limited in the range 10 - 20 cP³⁴.

One of the potential application of stretchable electrode is on devices that conform reliably to human skin, providing optimal comfort and dynamic fit, as in the case of smart patches and electronic tattoos³⁶. Thus, thinner electrodes and thin-film deposition methods on different substrates, such as thin elastomers and hydrocolloids help reduce the overall thickness of the

stretchable device towards the so called "imperceptible electronics"³⁷. In other circumstances where mechanical robustness is more important, for example in cloth, thicker electrodes are required. As a result, different printing technology may be called for different purpose. While inkjet printing is limited by relatively low ink viscosities, aerosol jet printing can tolerate a much larger viscosity range, between 1 and 1000 cP. Unlike in inkjet printing where ink droplets formed from printhead are individually placed on the substrate and coalesce to form continuity, aerosol printing forces the ink to form a fine mist, or "aerosol" that is further transferred to the substrate in a flow of carrier gas (e.g. nitrogen). Atomising transducer, based on either pneumatic force (Venturi effect) or ultrasonic energy, are used to generate aerosol of ink droplets typically 1 to 5 µm in diameter. The mists are further surrounded by a guided flow of nitrogen carrier gas and fed towards the deposition head, where they are injected out in the presence of a second gas flow – the sheath gas (also nitrogen) that helps constrain the ink mists to a small area. The opening end of the tip is between $100 - 300 \,\mu\text{m}$ in diameter, however, the action of the sheath gas enables focusing of the ink in an area as small as 10 µm in diameter³⁸. Advantages of this technique include highly customised features, choice of wide range of materials, large range of permissible ink viscosities, and reduction in material waste. Meanwhile, the presence of sheath gas flow additionally provides a cleaning mechanism of the substrate during the printing process. A wide range of materials including polymers, biological materials, carbon nanotubes and semiconducting materials have been reported successfully printed using aerosol jet printing technique^{39,40,41,42}. Moreover, by combining two different atomisers operating in the same printing work, different inks can be mixed in the aerosol form, and load ration against each other can be adjusted to achieve ondemand functionalities⁴³. Table 1 shows a comparison between the printing techniques reviewed

Technique	Nozzle(µm) ^{34,44}	Ink Viscosity (cP) ^{33,34,44}	Resolution (μm) ^{33,44}	Maximum Speed (m/s) ^{33,38}
Inkjet	10 - 50	10 - 20	20 - 100	0.2
Screen	N/A	~100	>100	0.5
Aerosol	100 - 300	0.7 - 1000	5 - 30	0.2

Table 1 - Comparison of the basic characteristics between the three common techniques

Repurposing conventional manufacturing techniques for stretchable electronics

Patterning thin film metal electrodes or interconnects on elastomeric substrates can be done via established fabrication techniques namely: lift-off, wet etching, shadow masking and pattern transfer. The main advantage of using these methods is related to the possibility of relatively quick industrialization pathways towards commercialization, together with the use of conventional, often depreciated manufacturing facilities. However, when the unique physical properties exhibited by elastomers are taken into account, such as the relatively large coefficient of thermal expansion (CTE) and swelling characteristics in commonly used solvents, major fabrication challenges are observed^{45,46,47,48}.

Due to the significant mismatch in CTE between most common photoresists (PR) that are commercially available and the elastomeric PDMS substrate, cracks are generally formed since early photolithographic process steps, which pose serious concerns on the reliability and reproducibility of the method. In an attempt to mitigate the effects of PDMS expansion during baking steps, Patel *et al.* used an epoxy-based SU-8 as sacrificial mask for lift-off, as the negative resist is characterized by CTE values closer to PDMS⁴⁹. By slowing the temperature ramping process, it was possible to avoid thermal cracking of the SU-8 mask.

However, cured SU-8 is difficult to be removed by solvents, so mechanical peeling of the mask is required. This limits the range of patterns that can be obtained, and affects the overall yield and reliability. Guo *et al.* approached these issues with SU-8 lift-off obtained by introducing polyacrylic acid (PAA) as intermediate layer between the epoxy resist and PDMS⁴⁵. PAA can be removed by water and also serves as barrier to limit solvents from reaching the bulk PDMS to minimize swelling⁴⁸. Nevertheless, pattern distortions and microcracks were observed due to the large residual stress in the SU-8 film. Microcracks resulted from the differences in CTE remain unresolved to date based on known literatures. However, a recent work showed that these microcracks (typical width < 1 μ m) do not present major concerns when patterning silver nanowires which are too large to penetrate through ⁵⁰

Examples of gold patterning fabricated via wet etching have been shown by several authors. Meacham *et al.* had constructed an elastic multi-electrode array based on PDMS with the method described above⁵¹. Gold was deposited on the PDMS, patterned through photolithography and subsequently etched by gold etchant. In a similar study, Lacour *et al.* introduced chromium stacks in between the PDMS and gold layer to improve the robustness as the transition metal is commonly used as adhesive layers⁴⁶. However, the etching profile obtained by wet treatment typically result in undercut due isotropic nature of the chemical reactions⁵². An undercut is either advantageous due to the easy removal of resist at the end stage or undesirable as dense patterns might merge and supporting structure can collapse. More importantly, etchants used in the processes are typically toxic which can be a critical issue for biocompatible applications if not sterilized properly. Wet etching of electrodes is now rarely seen as means to fabricate electrodes on elastomers.

On the other hand, stencil technology provides an alternative way to pattern metal electrodes on elastomeric substrates. It involves placing a sheet of pre-defined pattern in front of the substrate, which allows metal vapour to only pass through the pre-defined patterns. Minev *et al.* had patterned gold on PDMS via a Kapton[©] shadow mask⁵³. Similarly, Gong *et al.* deposited gold and titanium films through a shadow mask to fabricate interdigitated electrodes⁵⁴. However, the main limitation of this method is the relatively low resolution (>100µm) achievable which can potentially hinder the technological progress especially in imaging and recording systems based on stretchable materials^{55,56}.

More recently, researchers have developed techniques based on pattern transfer which not only mitigated the stated problems in lift-off but also avoided the direct exposure of elastomers to toxic chemicals. For example, Byun *et al.* had devised a method which first involve fabricating electrodes on silicon via standard microfabrication techniques and subsequently transfer the patterns to PDMS via (3-mercaptopropyl)trimethoxysilane as adhesive⁵⁷. In another approach, Deng *et al.* had also transferred electrodes made on a rigid substrate through carrier substrates such as poly(methylmethacrylate)⁵⁸. The downsides of these methods typically include the additional processing steps required, difficulties when handling the carrier substrates and also loss of patterns during transfer step(s).

Conclusions

Electronics that are highly stretchable and conductive have been demonstrated through efforts in material innovations and design, supported by both existing microfabrication technologies and also printing techniques. The field continues to attract strong interest from both the scientific and business community due to unmet needs that remain unsolved by conventional, silicon-based, electronic manufacturing and materials used by semiconductor industry. In this paper, we reviewed the enabling technologies required for stretchable electronics that are compliant with industry standards in terms of yield (90%), mechanical robustness (under repeated bending and stretching cycles, e.g. 10000-100000 cycles with less

than 5% degradation in performance) and reliability (i.e. under environmental and operational stress), as needed to enable commercialization of high-end applications that are at the frontier between bio-electronics and healthcare. Examples of such applications include smart prosthetic skin, implantable organ interfaces, "soft robotics" and "imperceptible electronics" where mechanical compliance is a key requirement in order to help the electronic system interact more naturally with living systems that are intrinsically soft and malleable. Using the enabling technologies discussed in this review, both emerging and existing electronic device manufacturers and system integrators are in the position to develop and commercialize new classes of products that address the reported manufacturing challenges of stretchable electronics, as required by applications in the energy, medical and military realm.

Acknowledgements

E.T is grateful for the studentship funding from Trinity Hall, University of Cambridge. E.T and L.O acknowledge funding support from the UK Engineering and Physical Sciences Research Council through the EPSRC Centre for Innovative Manufacturing in Large Area Electronics (Grant no. EP/K03099X/1). S.K-N is grateful for financial support from the European Research Council through an ERC Starting Grant (Grant no. ERC-2014-STG-639526, NANOGEN). Q.J is grateful for financial support through a Marie Sklodowska Curie Fellowship, H2020-MSCAIF-2015-702868. M.S is grateful for studentship funding from the Cambridge Trust.

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