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From Stretchable to Reconfigurable Inorganic Electronics

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Abstract: *Today's state-of-the-art electronics are high performing, energy efficient, multi-functional and cost effective. However, they are also typically rigid and brittle. With the emergence of the Internet of Everything, electronic applications are expanding into previously unexplored areas, like healthcare, smart wearable artifacts, and robotics. One major challenge is the physical asymmetry of target application surfaces, which often cause mechanical stretching, contracting, twisting and other deformations to the application. In this review paper, we explore materials, processes, mechanics and devices that enable physically stretchable and reconfigurable electronics. While the concept of stretchable electronics is commonly used in practice, the notion of physically reconfigurable electronics is still in its infancy. Because organic materials are commonly naturally stretchable and physically deformable, we predominantly focus on electronics made from inorganic materials that have the capacity for physical stretching and reconfiguration while retaining their intended attributes. We emphasize how applications of electronics dictate theory to integration strategy for stretchable and reconfigurable inorganic electronics.*

Keywords: Stretchable, reconfigurable, electronics, organic, hybrid, inorganic, mechanics.

1. Introduction

Electronics are key enablers for today's globally-connected digital world. Everyday we benefit from the pervasiveness of electronics, from computation and communication, to biomedical instruments and automobiles. Moore's Law has guided the Integrated Device Manufacturers (IDMs) community to physically scale transistors for performance improvement, increased functionality, and to make more affordable electronic gadgets. With the rise of mobile devices from the mid-nineties, society has grown accustomed to smaller, lighter and faster electronics. Continuous scaling in today's digital world has been possible due to the reliable batch manufacturing process based on complementary metal oxide semiconductor (CMOS) technology. In conjunction with the World's most abundant and affordable semiconductor material, silicon, CMOS technology is driving the digital world. CMOS electronics will continue to dominate for years to come. Still, a pragmatic shift in their form factor through physical flexibility and stretchability would enable further opportunities for applications where physically reconfigurable electronics are needed. This is essential for integrating electronics into an Internet of Everything, for instance, brain machine interfaces used for chronic monitoring of neurological phenomena, wearable skin patches for continuous monitoring of body vitals and drug delivery, or stimulation induced actuation (*i.e.* heat, light, and sound). In all these instances, adhesion between devices and soft tissue is prevented by the rigid and bulky state-of-the-art electronics. Inflexible devices are non-conformal, thus inhibiting effective interfacing, making them unsuitable in or on the human body, where joints go through various degrees of stretching, twisting and contraction. Hence, applications in advanced healthcare, such as thermal patches and vital monitors, require flexibility and stretchability, an adaptable reconfiguration is an important aspect in future

electronics. With such interesting and impactful prospects, the research community has significantly invested in the development of stretchable electronic materials. Obviously, organic and molecular electronics became the first attraction (covered in Section 2). Although naturally flexible and stretchable, they exhibit limited performance, thermal instability, and integration complexity, which has led to further exploration in 1D nanowires, nanotubes, nanorods and 2D atomic crystal structure based composite materials (covered in Section 3). Still, hybrid elastomers suffer from uncertainty in their properties, non-linearity, and integration complexity. Therefore, innovative engineering approaches have merged fractal architectures with classically established thin films like silicon, and III-V semiconductors, to transform them into stretchable and reconfigurable materials (discussed in Section 4). However, their brittleness poses significant issues for their use in stretchable electronic devices. In this review, we explore current techniques in flexible and stretchable materials development, their integration strategies and device characteristics under mechanical deformation. In the last section (5), we provide our perspective on the status-quo and vision regarding the future. Stretchable electronics are the epitome of engineering challenge, offering valuable opportunity in the context of material discovery and process innovation. In this review, we celebrate that by providing an accurate description of the status-quo, associated challenges and prospects for groundbreaking research.

2. Stretchable Organic Substrates

When we think of stretchable and flexible materials, the first thought that comes to mind is traditional polymeric or plastic materials. Naturally (proteins) or artificially produced monomers ^[1] are the building blocks of polymers, which compose organic materials—

materials that contain carbon [2]. Synthetically developed organic polymers and smaller molecules or monomers with a particular, controllable electrical behavior are the focus of this section. While these materials and the strategies involved in building organic/polymeric electronics can be advantageous mechanically (e.g., exhibiting flexibility and/or stretchability) and economically (low cost materials and tools for simple manufacturing processes), their electric performance is inferior to inorganic-material-based devices [3]. Nevertheless, organic materials and organic electronics have found important application to commercial niches, such as the display industry [4] and more recently the lighting industry [5]. In the following subsections we will review some relevant properties of organic materials, focusing specifically on their polymer building blocks, as well as a variety of fabrication strategies used to build organic-material-based electronics. Finally, we will discuss specific applications of organic/polymeric materials in electronics.

2.1. Intrinsically Elastic Polymers

The organic molecules that are used to develop organic electronics may be built from small molecules or much larger polymers. In this section, we will briefly describe the principles behind the mechanical properties of polymers, and we will review several fabrication processes currently used to build electronic devices from these polymers.

2.1.1. Basic Mechanical Properties of Elastic Polymers

As we have mentioned previously, one of the main features of most polymeric materials is their inherent mechanically flexible and often stretchable nature. The elastic subset of polymers, known as elastomers or rubbers, display a distinctive property of enables reversible deformations: this property is known as viscoelasticity. A viscoelastic material

exhibits both viscous and elastic characteristics when under deformation conditions [1]. The main difference between viscoelastic and purely elastic materials is evident from the hysteresis effect, which can be observed in the stress-strain curves of viscoelastic materials. In fluids, viscosity (resistance to flow) yields energy loss due to friction in the form of heat, and strain rate describes the rate of changing deformation of a material with time. Therefore, a viscoelastic polymer will dissipate energy in the form of heat when a load is applied to cause deformation (viscoelastic creep) and then removed (loading cycle) [6]. The area of the hysteresis loop represents this energy lost during a loading cycle. Microscopically, a polymer exhibits its viscoelasticity as molecular rearrangements. A creep will occur on a polymer subjected to stress and translates into rearrangement of the polymeric long chains that thus creates a back-stress in the material. Once the applied stress is removed, the polymer will return to its original form thanks to the accumulated back stresses [6]. We can think of it as the prefix “visco-” coming from the creep in the polymer, whereas the suffix “-elastic” from the full recovery of the polymer’s shape and length. A direct implication of these characteristics is a very low elastic modulus (known as Young modulus) and a higher fracture or rupture strength in the stress-strain mechanics compared with other materials. **Figure 1** shows the stress-strain curve of a typical polymeric material compared to a brittle material: a polymer accumulates much less stress with much higher deformation, whereas brittle materials deform and fracture very quickly.

2.1.2. Fabrication Processes

The diverse and versatile nature of polymeric materials makes several techniques for processing these materials into electronic devices viable options [7]. In general, the difference

between techniques is how the polymers are deposited onto the substrates and materials that will form the devices. These techniques range from spin-coating solution-based polymers to ink-jet printing [8, 9]. Patterning can be achieved through thermal transfer [10, 11], nanoimprinting [12], lithographically induced self-assembly [13], selective photo cross-linking [14], and cold welding [15, 16], among others [7]. One of the most common deposition methods is to coat polymers by solution processing. Once the solvent evaporates, a thin film uniform in thicknesses remains. Although this process is simple and can cover a large area, the solvents can be incompatible with or affect the layers beneath the polymer, thus impacting the performance and achievable complexity of the device [17]. As an alternative, compound polymers can be produced that are able to perform a variety of functions within a single layer [18]. Moreover, patterning and achieving complex structures with different materials and thicknesses becomes very challenging using only coating techniques. Ink-jet printing technologies can be more advantageous in this regard because polymers can be patterned micro-sized precision directly upon deposition [9]. Alternatively, thermal transfer, a dry-patterned deposition technique, involves taking the pre-deposited polymer material from a donor and selectively transferring it onto a receiving substrate through ablation by use of a heat source such as a laser [11]. Higher complexity can be achieved through this technique, although performance may be impacted due to thermal degradation during the process itself. Finally, metal-coated polymer films can be embossed (micro-cut) using a simple solid-state technique that forms metallic structures on polymer substrates with good resolution (down to micrometer sizes). More complex devices with higher performance are attainable using this method [19].

2.2. Devices, Challenges and Applications of Organic Electronics

Organic materials are universally acknowledged for their essential role in the development of display technologies that are commercially ubiquitous. The development of organic light-emitting diodes (OLED) dates back as early as the 1950s and 60s, when the mechanisms of organic electroluminescence were first described [20, 21]. This led to the development of the first practical, efficient OLED device in the late 80s, which triggered the rise of research and manufacturing of organic-based displays for use in devices ranging in size from handheld to big screens and monitors [22]. Despite their success in the display industry, logic applications have yet to demonstrate a performance advantage competitive enough to warrant commercialization [3]. A standard figure of merit for electronic performance is given by the charge mobility value in each material or device. In particular, organic materials have much lower mobilities than do inorganic materials, like silicon, the flagship material of the semiconductor industry, which has paved the way to today's era of information. Because silicon is a brittle material, current standard technologies based on it are rigid, planar, and inflexible unless some form of architectural or structural engineering modifies them at the system level. Where flexibility is concerned, organic materials have the upper hand because of their superior inherent mechanical elasticity. The highest mobilities in organic materials have been achieved with semiconducting thin films in the order of 10^1 cm²/Vs (two orders of magnitude below silicon); for example, the mobility of an organic thin film transistor (OTFT) based on a highly aligned, meta-stable C8-BTBT:PS crystal spin coated on polystyrene using a novel off-center method reached up to 43 cm²/Vs [23]. Additional examples of organic-based devices with high mobilities include the work by Podzorov *et al.* in 2003, who demonstrated a mobility magnitude of 8 cm²/Vs by optimizing the fabrication

process of rubrene-based single-crystal organic field-effect transistors [24]. A decade later, Wei Xie *et al.* showed a transistor based on rubrene-d₂₈ single crystals with a consistent mobility of 10 cm²/Vs at room temperature [25], and in the same year, Il Kang *et al.* built an OTFT with a mobility of 12 cm²/Vs through side-chain engineering of two polymer semiconductors (P-29-DPPDBTE and P-29-DPPDTSE) [26]. Note that predominant organic-based devices are p-type and thus the reported mobilities correspond to holes; electron mobilities are usually several factors lower (the highest electron mobility of 6.3 cm²/V.s reported was achieved with the donor-acceptor polymer PDBPyBT by Bin Sun *et al.*) [27]. To develop complementary metal-oxide semiconductor (CMOS) logic, the mobility of electrons and holes need to match, a continuing challenge in organic electronics. Furthermore, many organic-based transistors that show high relative mobilities have the evident disadvantage of high sensitivity to air or the need for complex processing conditions [28-30]. Researchers are developing strategies to produce more stable compounds, such as the one demonstrated by Jun Li *et al.*, which is based on an ambient-stable, solution-processed DPP-DTT polymer, which exhibited a hole mobility up to 10.5 cm²/V.s [31]. Two remaining challenges worth mentioning include high operation voltage and the device scalability that are necessary for increasing the integration density (increasing computing power and functionality). Silicon-based devices have reached a level of ultra-large-scale integration (ULSI – billions of transistors in an area of about 2 cm by 2 cm), whereby at present, transistor gates of only 14 nm in size are produced; this is unquestionably a very high ambition for organic-based electronic systems.

Still organic electronics have the potential for a variety of applications beyond displays and flexible logic devices. For one, their manufacturability may allow the development of large-

area organic macro-electronics [8]. The promise of low-cost fabrication of large-area, flexible electronics could be delivered through promising new solution coatings and roll-to-roll techniques. For example, Ying Diao *et al.* developed a method for fast coating and patterning of cm-long, single-crystalline organic Pentacene-based thin films to produced devices with mobilities in the range of $\sim 8 \text{ cm}^2/\text{V.s}$ [32]. And for two, the compatibility of many of these organic materials with biological specimens has led to the emergence of bioelectronics, a subject of current scientific interest. The elastic properties of organic materials make them optimal for applications to either stimulate biological tissues/muscles or to transmit biological signals [33]. For instance, Simon *et al.* developed an organic electronic ion pump that could precisely deliver several neurotransmitters both *in vitro* and *in vivo*. This device selectively stimulated nerve cells in response to specific neurotransmitters [34]. Later on, Campana *et al.* demonstrated a conformable organic electrochemical transistor (OECT) with micro-patterning of PEDOT:PSS, fabricated on a resorbable, bio-scaffold substrate for electrocardiographic recordings of cardiac muscle. The OECT, which was in direct contact with the physiological sample, displayed promising results with high sensitivity, responded quickly and had high bending stability [35].

3. Stretchable Hybrid Materials

Typically, materials that have high conductivity and stability tend to have poor mechanical robustness and elasticity while more flexible materials tend to have poor electrical properties. Therefore, extensive research has been dedicated to developing materials that are both stretchable and highly conductive for application to wearable technology. Some initial work towards this goal began by fabricating stretchable electronics on polymeric

substrates, such as polydimethylsiloxane (PDMS), polyethylene terephthalate and polyethylenimine (**Figure 2a**). The mechanical elasticity of these polymers suppresses strain localization on the relatively rigid metal layers, leading to the realization of bendable and stretchable devices for applications in stretchable displays, artificial skin, monitoring systems and bio-integrated devices [36]. However, the deposition of metal interconnects on top of polymeric substrates has its limitations: this technique gives rise to three possible modes of failure, such as slipping, cracking and delamination, seen in **Figure 2b** [36]. For example, elastic breakdown of metal films under only a small applied strain can cause microcracks that propagate in the metal eventually cause electrical discontinuity and delamination results from an accumulation of strain stress on the rigid metal conductor [37-39]. Ductility of the device is limited by the thickness of the rigid conductive film and stretchability is limited by separation of the metal grains toward total electrical discontinuity. Thin films can sustain some strain before fracture, but their strain endurance can be improved using pre-straining deposition techniques and buckling. Nevertheless, weak adhesion and large modulus mismatch between the polymer substrate and the conductor film render these approaches to improve strain tolerance insufficient for producing highly stretchable devices. Further details about the challenges of these approaches are available in previous reports [40-47].

Figure 2c depicts some of the strategies that have been applied to advance the capacity of stretchable hybrid conductive elastomers. Combining materials to produce hybrids of polymers, such as silicone rubbers, with electrically conductive nanoparticles, metal nanowires (NWs), graphene sheets and carbon nanotubes (CNTs) has been a successful approach [41, 48-52].

In this section, we will discuss some of the popular approaches used for preparing hybrid elastomers, their challenges and their distinct advantages for a specific set of applications. We will list strategies used for enhancing the electrical and mechanical properties of stretchable conductors and present some of the recent applications dominating this field. From this is a huge area, where a variety of nanomaterials can be used for constructing hybrid elastomers, we focus only on the latest findings using NWS, CNTs, some graphene derivative composites and liquid metals and ions.

3.1. Stretchable Hybrid Conductors

Hybrid elastomers are the most common choice of conductor used to overcome several unresolved challenges in stretchable electronics, including (1) device stability under high strain conditions, (2) simultaneously achieving high conductivity, high mechanical robustness and excellent stretchability, (3) attaining high transmittance along with high electrical conductivity for optoelectronic applications and (4) improving the interfacial interaction between the device material and the carrier.

3.1.1. Carbon Nanotube Elastomer Composites

Carbon nanomaterials such as carbon black and CNTs have fascinating properties for application as composites. CNTs are known for their robustness and advantageous electrical properties and have been extensively used in conjunction with polymeric materials to make stretchable conductive composite mixtures. Mixing elastomers with fillers (*i.e.*, conductive composites) can significantly improve the mechanical and electrical properties of polymers. Carbon black is the most commonly studied and used filler of elastomers [53-57]. However, carbon-black-based hybrids often require high fractions of carbon black to improve the

rubber's conductivity and strength, and often demonstrate coarse aggregation and agglomeration of particles in the matrix network, obstructing the conductive path [53]. Thus, research is shifting toward more eco-friendly alternatives, such as CNT composites, which possess high intrinsic electron mobility and intriguing mechanical properties [58, 59]. The use of CNTs as fillers reinforces stretchable polymers and adds desired electrical functionalities [60-65]. For example, Takao Someya *et al.* demonstrated that single-walled carbon nanotubes (SWCNTs) make a highly conductive filler [66]. SWNT composite films were prepared by suspending the CNTs in a fluorinated copolymer. This is achieved through the uniform dispersion of SWNTs in an ionic liquid 1-butyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide (BMITFSI), and the generated mixture can be processed as a paste-like material (**Figure 3a**). The SWCNT-based mixture can then be sandwiched in between two cured layers of PDMS for stability. Electrical conductivity and mechanical properties can be changed by altering the weight percent (wt. %) of SWCNTs in the mixture to a maximum of 20 wt. % without substantially degrading the stretchability of the polymer. The resulting stretchable conductor exhibits a conductivity of 57 S.cm^{-1} and a stretchability of 134%. Electrical stability is only reported up to 38% of uniaxial stretching with a decrease to 6 S.cm^{-1} upon an applied strain of 134% [66]. This degree of electrical conductivity remains insufficient for operating an integrated circuit, but compared to commercially available CB-based rubbers, which display a conductivity of 0.1 S.cm^{-1} up to 160% tensile strain, this work shows orders of magnitude improvement in electrical conductivity. Other similar works have involved preparing a hybrid composite using multi-walled carbon nanotubes (MWCNTs) dispersed in PDMS mixture and then embedding micropatterns of the conductive ink into bulk PDMS to form stretchable devices [51]. Achieving monodispersion of MWCNTs

within PDMS or any other polymer can be challenging due to the aggregation of individual CNTs, resulting in the formation of clusters that inhibit current transfer. To prevent this problem, an organic solvent, such as toluene, is often used to dissolve the base polymer more easily, such that the decreased viscosity of the polymer mixture enables the formation of a homogenous network of MWCNTs within the matrix. Following this method, PDMS base polymer is added to toluene (1:4 volume ratio) and MWCNTs are separately dispersed in another toluene solution (5 wt. %). The two solutions are then mixed together using magnetic stirring until all volatile toluene has evaporated under the applied heat [51]. A PDMS curing agent is then added to the mixture to prepare a hybrid MWCNT/PDMS conductive printing ink. Although this method has advantages for ink micropatterning, the reported electrical conductivity of this mixture remains very low, up to 6 S.cm^{-1} for a maximum of 15 wt. % of MWCNTs. Furthermore, piezoresistive polymer nanocomposites are highly desirable for stretchable mechanical sensing applications. MWCNTs/elastomeric triisocyanate-crosslinked polytetrahydrofuran (ETC-PTHF) nanocomposites have recently become an attractive material because of their high stretchability of up to 700% and high sensitivity to mechanical stimuli [67]. CNT/ETC-PTHF nanocomposites are prepared by mixing p-methoxyphenyl-functionalized CNTs and ETC-PTHF under ultrasonication. Again, electrical conductivity of the resulting material increases with the ratio of CNTs. However, electrical conductance of the stretchable film decreases dramatically by 42,455 times with an increased applied strain up to 500%. Nevertheless, the developed piezoresistive hybrid composite has applications in artificial skins [68], wearable health monitors [69] and electronic textiles [70]. Kim's group have recently developed an improved technique for achieving robust and durable conductive elastomers by producing stretchable conductors that are three times

less stiff and 45 % more electrically robust to physical deformations than previously built CNT/PDMS elastomers, by integrating air bubbles into the elastomer (Figure 5a) [71].

Although these processes for building CNT composites have potential for some sensing applications, the products lack the conductivity and stability required for several other applications. Thus, different techniques and composite materials have been used as an alternative to CNTs to provide optimized performances, targeted at improving functionality and stability for a diverse set of applications.

3.1.2. Metal Nanowire Elastomers

The realization of stretchable electronics generally requires the development of highly conductive, stable, stretchable conductors that have application not only for sensing purposes, but also as a mesh of interconnects for the whole circuit. The intrinsic properties of NWs placed them in the spotlight for their capacity to solve the challenge of preserving high conductivity while maintaining high mechanical stability and elasticity in stretchable electrodes. Metal NWs exhibit omnidirectional flexibility due to the ultra-thin nature and advantageous stretchability properties that arise from their singular dimensionality (1D). Only 1D NWs can achieve this high stretchability through the percolating network design, explained by the percolation theory [72, 73]. The resultant excellent electrical and mechanical properties of metal NWs have made them an attractive option for use in stretchable conductors. In general, 1D nanostructures are good candidates for conductive interconnects in soft electronics because the 1D materials have a strong tolerance to stretching as a result of intersliding behavior [51, 64, 74].

Given the advantages of using NWs in stretchable conductors, stretchable electrodes are prepared using the same coating techniques used for flexible electrodes. Commonly, the deposited composite film undergoes transfer or embedding processes because of the poor adhesion of NWs with polymers. In an embedding technique, a network of NWs is sandwiched in between two polymeric layers. This method allows for a more uniform stress distribution profile under applied strain. Usually, direct stress applied on bare NWs leads to discontinuity in the network and a sudden increase in resistance. In contrast, embedded NWs can disperse the stress since strain localization is suppressed by the polymer substrate [75]. In this case, the main preparation methods used are drop-casting and dip-coating techniques, as shown in **Figure 5b** [76, 77]. In a recent study, Lian Gao *et al.* demonstrated ultra-stretchable composite fibers based on silver nanowires (AgNWs) through a dip-coating process [50]. The hybrid fiber possesses a high conductivity up to 4018 S.cm^{-1} , and maintains a high conductivity of 688 S.cm^{-1} at 500% tensile strain. The final structure of AgNWs is embedded in a thin PDMS layer using dip coating to mechanically strengthen the structure, as shown in **Figure 3b**. The work correlates the dip-coating process time to obtained conductivities, where increased dip-coating times lead to a three time increase in conductivity.

Although the high conductivity along the axis of NWs makes them preferred candidates for stretchable conductors,, challenges, such as surface roughness and high contact resistance, remain a threat to efficiency.

3.1.3. Mixed-Metal Composites

In general, a small composite diameter, a large young's modulus, a high yield strength and a high tensile strength are considered properties that are desirable when building stretchable electrodes [78, 79]. Nevertheless, the degree of adhesion to polymeric substrates by nanostructures determines the mechanical stability of the conductive elastomer. Although NWs have been used extensively in stretchable electrodes, defects and dimension and adhesion issues affect the strength of networks, and can result in weakened mechanical and electrical properties. To overcome these challenges, hybrid composites that incorporate metal nanocomposites inside polymers have been developed. Mixing NWs with another conductive nanomaterial became a solution for overcoming the limitations of film conductivity and sheet resistance incurred by using NWs alone [80, 81]. Most NW hybrid systems target adhesion optimization between NW and polymer, toward improved mechanical stability. Examples of mixed-metal NW hybrid composites and strategies to improve stretchability of conductors can be found in a review paper presented by *Zhu et al.* [43]. For instance, mixing AgNWs with SWCNTs increases elasticity and improves adhesion, leading to mechanical stability under 460% strain, and enhances the stretchability of the AgNW network up to 480% strain [82]. CNTs fill the space between nanowires and thus reduce deformation of the NW network. This leads to a decrease in network resistance due to locally developed electron transport paths. A 2016 study by Soon Hyung Hong *et al.* demonstrated that in CNT-Graphene-PDMS hybrid fillers, the synergistic effects in the CNT-graphene mixture prevent agglomeration of CNTs and the restacking of graphene to improve electrical performance while maintaining high stretchability [83] (**Figure 3c**).

3.1.4. Graphene-Conjugated Composites

A graphene hybrid composite has superior mechanical properties over NW-based strategies. Despite the high stretchability of NWs evidenced by the percolation theory, their intrinsically low mechanical strength and low adhesion with polymers limits their use for highly stretchable conductors. Graphene has better adhesion with polymers due to strong Van der Waals interactions. Graphene sheets enable improved homogenous dispersion in the polymer matrix, leading to enhanced mechanical and electrical properties of graphene-polymer conjugates. Yibin *et al.* show the use of a graphene-polyimide nanocomposite foam translates to high mechanical flexibility and superelasticity when applied to strain sensors [48]. The reduced graphene oxide/polyimide (rGO/PI) nanocomposites were fabricated using freeze casting and thermal annealing. The hybrid elastomer displays desired electrical conductivity, high elasticity and excellent stability due to the synergistic effects between rGO and PI. This conductive foam is highly compressible, exhibiting linear behavior up to 70% strain with no observed hysteresis effect, and causing it to gain in popularity for use in both pressure and strain sensors [48]. More recently, Ali Khademhosseini *et al.* integrated GO nanoparticles into a hydrogel elastomer to engineer a biocompatible and stretchable hydrogel with tunable electrical and mechanical properties [84]. The result was an electrically conductive and highly elastic MeTro/GO hybrid hydrogel, where methacry-loyl-substituted tropoelastin (MeTro) is a biocompatible elastomer (Figure 2d). Previous attempts to produce conductive hydrogels using gold nanowires (AuNWs) [85] and CNTs [86] have also been demonstrated, but the outcome involved significantly degraded elasticity of the elastomer [53]. For the development of biocompatible stretchable devices, GO/hydrogels have

excellent potential for application to tissue engineering and regenerative medicine applications.

3.1.5. Liquid Metals and Ionic Liquids

Liquid metals in polymers are an interesting field for the development of stretchable conductive pathways [87]. Some metals and metal alloys have very low melting points below or around room temperature. Some examples include mercury, eutectic gallium–indium alloy (EGaIn), gallium–indium–tin alloy (Galinstan), and just recently, the biphasic Ga₂Au alloy [88]. As liquids, these metals have gained significant interest because of their high electrical conductivity ($1.0 \times 10^6 \text{ S.m}^{-1}$ for Hg and $3.4 \times 10^6 \text{ S.m}^{-1}$ for gallium–indium alloys) and extreme stretchability, > 600 %, without losing conductivity [89, 90]. As an alternative to the toxic mercury-based stretchable conductors that have been heavily investigated in the past [91, 92], EGaIn and Galinstan have received more attention recently [93-95]. Whitesides *et al.* presents some examples of stretchable microfluidic channels filled with EGaIn [95, 96]. Other groups have also fabricated liquid-metal patterns through screen-printing and embedding techniques [97, 98] (Figure 4a), where electrical resistance changes with mechanical deformation of the liquid metal upon stretching. Although the fluidity of liquid metals provides durability under large applied strains, it can be also seen as a limitation, especially for micropatterning when detailed features would not be properly resolved. Majidi *et al.* studied high-density micropatterning of liquid metals by pressing an elastomer mold onto an EGaIn film [94] to prevent any expansion of the metal after patterning. Note that liquid metals tend to lose their stretchability at temperatures above their melting point, which can be as low as 15.5 °C for EGaIn [99], and thus restricting their use for some

applications. In addition, when liquid metals are highly stretched, the oxide layer on their surface breaks, resulting in poor electrical stability under stretching cycles [94]. Moreover, liquid metals adhere poorly to their polymeric substrates due to their large surface tension (hundreds of mN.m^{-1}) [100]. Thus, further progress and advances are necessary to overcome the many challenges accompanied with the implementation of liquid metals in stretchable devices.

As an alternative to liquid metals, ionic liquids can also be incorporated into polymers. A liquid-wetting-solid method has been under investigation for its potential to overcome challenges commonly associated with the mechanical mismatch of stretchable conductors, such as material delamination and local fracturing under large strains. This method was recently presented by Wenlong Chen *et al.*, who integrated ionic liquids as the conductive components with polymers to produce stretchable conductors on fabric (Figure 4b) [101]. While liquid metals display a high surface tension that limits their scope of application, conductive ionic liquids adhere more firmly to flexible polymers. In their simple approach, Ma *et al.* successfully turned a variety of soft elastomeric supports into stretchable conductors. Their ionic-liquid-based conductors display exceptional performances at ultra-large strains ($\epsilon > 600\%$) with high sensitivity down to a low strain of 0.05% [101]. Using the technique shown in Figure 4b, the group built ionic-liquid-based piezoresistive sensors that were capable of attaching to human skin and of being integrated into clothing to detect human motion. The strategy of using soft liquid materials to generate stretchable conductors seems to be a promising approach for producing high-performance stretchable sensors. Furthermore, the concentration of ionic liquids can be varied and other conductive materials can be incorporated into devices to tailor the molecular structures and tune the ionic

conductivity [102, 103]. Although ionic liquids possess the advantage of lower surface tension and a lower Young's moduli than previously reported approaches, further improvements are necessary to reduce the megaohmic resistances they produce that are not favorable for many integrated electronic devices.

3.2. Electrical, Optical, and Mechanical Tuning of 1D Materials

Having reviewed the advantages of stretchable conductive materials and many of the techniques used to overcome their limitations, we will now focus on the main challenges originating from 1D composites and the techniques used to tune their electrical, optical and mechanical properties, optimizing their use for a targeted application.

3.2.1. Percolation Theory

The percolating theory plays an important role in determining electrical, optical, and mechanical properties of 1D materials [104]. For 1D wires, the percolation theory states that the resistance, R_s , of the formed cluster is related to the wire's average length, L_s , density, D , and diameter by Equation (1) and Equation (2), demonstrating that the conductivity of a NW network, for example, can be enhanced through the elongation of the NWs [49, 105]:

$$L_s \sqrt{\pi N_{th}} = 4.236 \quad (1)$$

$$R_s \propto (D - N_{th})^\alpha L_s^\beta \quad (2)$$

Where N_{th} is the threshold areal wire density, α is a parameter related to spatial arrangement, and β is a parameter related to junction resistance and intrinsic conductivity of the material. Equation (2) describes the sheet resistance of the developed network. The

percolation theory equations clearly show that the percolative behavior of 1D materials increases the conductivity of the NW network for NWs that are longer, smaller in diameter and exhibit a high density within the network formed. However, if the network contains NWs that are too small in diameter, then the mean free path of electrons in the material's bulk increases, dominating the percolation theory and leading to an increase in the network's resistance.

Vacancy, hence density in the network of NWs, determines the transparency of the film because transmittance loss is mainly induced by scattering of light within the 1D shape of NWs. Haze can be controlled by the density and diameter of NWs: smaller diameter and longer length NWs reduce haze. For stretchable hybrid materials, a greater vacancy is desirable because it minimizes the light scattering effect for a more transparent film. In addition, the degree of vacancy is strongly dependent on the coating and post-processing methods used to achieve desired optical effects for optoelectronic applications [106-118].

3.2.2. Dispersion in Surfactants

1D materials, such as NTs and SWNTs, dispersing in aqueous surfactant solutions have been found to lead to higher percolation threshold, N_{th} , which translates into a decrease in the resistance of the network compared with non-dispersed composites [119]. One of the main reasons behind this phenomena is that the surfactant prevents intermolecular interaction between the NTs, whereas in the absence of a surfactant, the NTs adhere strongly through Van der Waals forces, resulting in an increase in the effective average length of NTs in the network.

3.2.3. Alignment

The alignment of NWs can have a major impact on the resistance of the network [120]. Two conditions are primarily responsible for a decrease in the network's conductivity: (1) an abundance of junctions between NWs and (2) a small contact length between consecutive NWs. Typically, electrical alignment is applied to improve the alignment of NWs through dielectrophoretic forces in an alternating electric field. The electric field that is generated induces charge separation, and the resulting polarization generates a dipole moment, which aligns the NWs into an energetically favorable orientation parallel to the field lines [121]. The dielectrophoretic force F can be expressed by Equation (3):

$$F = \frac{1}{2} \alpha v \nabla |E|^2 \quad (3)$$

Where α is the effective polarizability of the structure, E is the electric field strength and v is the volume of the 1D structure. Thus, the aligned configuration and percolated networks form electronic devices with robust interconnections and electrodes.

The alignment of the NW network affects not only the electrical properties but also the optical properties of the stretchable network/film produced. This optical change is due to a set of properties occurring at the nanoscale level, where optical properties in 1D structures are dominated by quantum confinement effects and are due to an increase in the surface area per volume ratio.

3.2.4. Resolving Junction Resistance

NWs and other 1D structures exhibit high resistance on the junction side due to quantum mechanical tunneling of electrons across stacked NWs. But, if surfactants are present on the

junction side, the tunneling of electrons from one NW to the other will be inhibited. This leads to an increase in junction resistance and thus an increase in contact resistance of the electrodes. There are several methods to address and reduce junction resistance. First, properties of the NW itself can control contact resistance and light scattering: the percolation theory explains that smaller diameter longer length NWs have higher conductivity and produce less haze. The welding technique is the best-known and most widely used method for decreasing contact resistance. This process involves the formation and solidification of a liquid phase at the interface [122]. It is initially prepared by fusing the NW junction through heat exposure. This can be done by pumping in extra energy, which will locally heat up the junction through the vibrations of electrons. The welding technique can be accomplished using direct heating [122], mechanical pressure [123] or plasmonic welding (**Figure 6a**) [124]. These processes join similar or dissimilar materials by causing a chain of phase change that leads to inter-diffusion followed by cooling. NWs in a stretchable matrix that have been tightly linked by welding will follow the flexibility of the substrate and be more mechanically stable. For instance, a physically welded NW mesh network shows the mechanical tendency of twisting, with stretchability up to 100% for a biaxial strain [125]. Meanwhile, chemical welding is considered to be a part of post-treatment processes with chemical compounds or the integration of NWs with other materials. For example, PEDOT:PSS, graphene, or ZnO can be used as a coating on top of a AgNW film, altering the behavior of the NW electrodes [126-128]. Choy *et al.* demonstrated a chemical welding process using a simple alcohol-based solution approach, where they weld crossed AgNWs by chemically growing silver solder at the junctions of the NWs (**Figure 6b**) [129]. This led to the successful development of transparent AgNW-based electrodes with improved electrical conductivity and stability.

By carefully evaluating the effect of NW parameters through model simulations, Karen Winey *et al.* found that low sheet resistance ($R_s \leq 100 \Omega/\text{sq}$) correlates with high optical transparency ($\%T > 90\%$) [130] (**Figure 6c**). They conclude that achieving lower contact resistance reduces the sheet resistance with the same number of NWs [130]. Their study demonstrates that modulating junction resistance is central to adjusting the surface roughness, electrical properties and optical properties of composite electrodes.

3.2.5. Improving Adhesion

One of the ways to tune the electrical and optical properties of stretchable conductive networks is through surface modification. As mentioned previously, adhesion of the NW layer to the substrate affects the surface roughness and homogeneity during the coating process, which plays a critical role in determining the mechanical, electrical and optical properties of the NW film. 1D metal NWs have a rough surface that can decrease the electrical performance of the device. Primarily, out-of-plane orientation of the wires cause poor mechanical adhesion to the substrate [131], leakage currents [132] and short circuits [133] in the system. Low adhesion causes friction on the NWs, leading to electrical failure when strain is applied. This mechanical failure can be fixed by adopting a strong adhesion, which can be developed by surface modification of the substrate or the composite material used (NWs, CNTs or graphene). Examples of common surface modification methods are plasma treatment [134] and chemical treatment [135].

Surface roughness can also have an impact on the optical properties of the material. Because light scattering is amplified by rougher surfaces, the haze factor increases, which is typically undesirable for transparency in optoelectronic applications. Nevertheless, Qibing Pei *et al.*

fabricated highly efficient OLED devices with only a few nm of surface roughness [136]. They report the fabrication of an elastomeric polymer LED comprising transparent stretchable electrodes made from a thin percolation network of AgNWs. They produce the device using an all-solution-based process that emits light when it undergoes strains up to 120 % establishes high transparency in the visible regime, has good electrical conductivity, high stretchability and high surface smoothness. The key to their success was using AgNW–poly(urethane acrylate) PUA composite electrodes, which have not only high transparency and excellent stretchability, but also the PUA polymer has strong adhesion properties with AgNWs, which translates into reduced surface roughness.

3.3. Applications of Stretchable Conductive Composites

Engineering of composite hybrid materials offer a wide range of applications in stretchable electronics, such as displays, touch screens, bio-integrated sensors, artificial skin, data storage and energy harvesting devices (Figure 7 and 8) [137]. In the field of optoelectronics, metal NW films are among the most promising alternatives for next-generation stretchable transparent conductors [49, 116, 124, 132]. For example, AgNW stretchable conductors are becoming preferable to other flexible candidates, such as indium tin oxide (ITO), for the fabrication of stretchable displays (Figure 7e and f) [129, 136, 138].

Hybrid conductors are becoming popular for the development of stretchable artificial skin, where strain and pressure sensors are commonly employed to detect bodily information such as muscle movement, heart rate, respiration, blood pressure and mechanical properties of skin. (Figure 7d and Figure 8c). Stretchability, stability and electrical performance are all important characteristics of a sensory platform that is suitable for a range of applications.

For stretchable conductors, these characteristics are tuned depending on their target function and conditions of application. Exploiting the characteristics of hybrid materials, such as conductivity, elasticity, transparency and piezoresistivity, has enabled the development of a variety of soft, stretchable sensors, including strain, pressure, tactile and acoustic sensors [40, 68, 69, 80, 84, 139]. For instance, carbon black composites in soft elastomers led to the development of wearable piezoresistive strain sensors for taking movement and assessing skin properties. Going forward, NW-based hybrids are preferable as artificial skin sensors because of their extremely high sensitivity and mechanical elasticity [40, 140, 141]. For example, a AgNW/PEDOT:PSS/PUA nanocomposite was used to build a highly sensitive, stretchable, transparent platform for a self-powered skin strain monitoring system (**Figure 8c**) [142]. 1D carbon nanomaterials are also used as an alternative to NWs in artificial skins because of their greater sensitivity to external stresses and superior mechanical capacity for bending, stretching and twisting compared to conventional pressure-sensitive rubbers. Zhenan Bao *et al.* produced stretchable, transparent SWNT-based pressure and strain sensors (**Figure 7a**) [143] to support the uniform integration of sensor arrays over large surface areas, and a chameleon-inspired stretchable electronic skin was developed using an interactive tactile sensing platform constructed from pyramidal-micro-structured SWNT-coated PDMS films (**Figure 8a**) [144]. Moreover, the transparency or semi-transparency of engineered stretchable conductors are enabling the development of new classes of interactive artificial skins for epidermal optoelectronic applications [145]. The benefits of hybrid conductors are also being applied to thermal and electric actuators for developing skin-based therapy applications: dielectric elastomers, electrostrictive polymers or liquid crystal elastomers are typically chosen as the active material for an actuator where the

elastomer changes shape in response to an electric field [146, 147]. However, when devices are actuated by the movement of ions, CNTs, NWs, conductive polymers or ionic polymer-metal composites are preferable [148]. For instance, D-H Kim *et al.* designed a soft stretchable heater from AgNWs and an elastomer (styrene-butadiene-styrene; SBS) composite targeted for articular thermotherapy (Figure 8b) [149]. The high conductivity of AgNW hybrids enabled the fabrication of RLC resonant circuits integrated with sensors for wireless monitoring [150]. Finally, stretchable, conductive composites have applications in energy supply devices and data storage devices. A stretchable piezoelectric nanogenerator was reported using a hybrid composite of microparticles with MWCNTs dispersed in elastomer (Figure 7b) [151]. And a floating gate memory was fabricated on a wearable platform using a soft SWCNT composite [152]. In data storage applications, the primary advantages of these stretchable hybrid conductors are their capacity for repetitive deformations and accumulated fatigue cycles.

4. Stretchability by design

Stretchable electronics can be obtained by taking stretchable materials and making them electronic or by taking electronic materials and making them stretchable. In the previous sections, we discussed about the inherent stretchability of some materials, including polymers with polymer chains that slide along the axis of the applied force, enabling strain without yielding. However, these materials have poor applicability to the semiconductor electronics industry due to their poor electrical, mechanical and thermal properties. In this section, we will discuss the materials used in the CMOS industry, such as silicon, silicon oxide, aluminum and copper. Although these materials have excellent electrical properties their application to state-of-the-art electronics is limited by their lack of stretchability [153, 154]. In

the case of some metals, such as gold, rearrangement of grain boundaries limits their stretchability. In the case of purely crystalline materials, the atoms in the lattice can be moved away from each other to provide stretchability. However, this requires very large forces that require a high modulus of elasticity (stress/strain) and a low yield strain. Patterning of crystalline and polycrystalline materials can allow the strain energy to be used to reconfigure the structure while the material itself undergoes negligible or no strain. Hence, high apparent stretchability can be achieved with low yield strain materials. This technique of patterning rigid/semi-rigid materials into stretchable structures is well known and is commonly seen in metallic springs [155]. In the following sections, we will investigate previously demonstrated works in stretchable electronics using patterned thin film materials.

4.1. Stretchable semiconductors

Although there are several benefits to crystalline silicon that have made it the material of choice for most state-of-the-art electronics to date, techniques that could impart stretchability to silicon are highly desirable. In 2007, Huang *et al.* presented a process for obtaining stretchable silicon [156] using both silicon (100) and silicon (111) substrates. Using silicon (100), a thick silicon-on-insulator (SOI) substrate with a 30- μm -thick top layer of silicon and a 400-nm-thick layer of buried oxide (BOX) was used. The top silicon layer was patterned in the form of lateral spring structures using deep reactive-ion etching (DRIE). The silicon was etched all the way to the BOX layer, and a protective layer of low-pressure chemical deposition (LPCVD) silicon oxide was conformally deposited. The deposited oxide and BOX layers were etched using magnetically enhanced RIE (MERIE) to expose the silicon

handler substrate. The spirals were then released using XeF₂, according to the following chemical reaction:

Using silicon (111), the spiral patterns were etched up to a desired thickness using DRIE and LPCVD silicon oxide was deposited to protect the sidewalls. The oxide at the bottom of the trenches was removed using MERIE and an extra DRIE step was performed to punch through the bottom silicon (111) plane. The structure was then subjected to 5% TMAH etching at 90 °C for 80 minutes to selectively etch the other silicon planes.

Although hydroxide ions do not appear in this reaction, the reaction rate is greatly increased by the presence of these ions. Furthermore, the presence of hydroxide ions helps in dissolution of the reaction product. This process was used to pioneer the research on patterned and released silicon substrates for use in stretchable electronic applications. A modification of the process was demonstrated by Rojas *et al.* in 2014 [157], whereby the top silicon layer of a thick SOI substrate was etched up to the BOX layer using DRIE and the BOX was removed using selective etching and hydrofluoric acid (HF) vapor according to the following equation:

The process flow schematics for both the processes and the optical images of the resulting silicon structure are as shown in **Figure 9**. These processes use a high aspect ratio etch to

define the spiral structure. When a lateral force is applied, the spirals unwind, providing stretchability. The work done in stretching is absorbed by the spiral as strain energy. When the force is removed, this energy is released, bringing the spring back to its original position. Thus, the silicon islands connected by the springs do not undergo strain. This is essential from an electronics point of view because strain in a silicon lattice alters its charge transport characteristics, thus potentially changing the performance of the circuit components. The stretchability obtained using this method depends on the width, radius and the number of turns of the spirals. Rojas *et al.* reported a 10-fold increase in lateral stretchability and a 30-fold increase in area stretchability [157].

In 2014, Kim *et al.* patterned and etched the top silicon layer of a regular SOI substrate (top silicon thickness = 80 nm) and removed the BOX layer using HF [158]. Their process used a different type of lateral spring structure—a serpentine or horseshoe spring. The fundamental difference between this and the previously shown structure is the deformation method. In Kim *et al.*, the spring undergoes twisting at the peaks and troughs to absorb the work done during straining. This design can be made using ultra-thin films with large lateral widths. Thus, the top layer of a SOI substrate could be used by implementing standard lithography and etching techniques.

4.2. Stretchable Metal Interconnects

One of the ways of obtaining stretchable electronics is to have small electronic islands connected with stretchable metal interconnects. This shifts the problem from having stretchable semiconductor to a stretchable metal. This is an easier problem to solve because electronic properties of metals (such as carrier transport) do not change significantly with

application of strain. Hence, a small amount of strain can be applied without appreciable changes to circuit performance. However, application of large strain can cause metallic thin films to crack and lose their conductivity. One of the ways of overcoming this was demonstrated by Sun *et al.* in 2015, wherein they deposited a copper thin film in a columnar structure [159]. In general, metallic interconnects can be designed in the form of stretchable lateral spring structures such that the strain energy is absorbed by the deformation of the structure and the strain experienced by the thin film is minimal. In the following section, we will look into some of the recently demonstrated stretchable metal interconnect designs, their fabrication processes and their application.

4.3. Designs and Applications

The lateral spring designs adopted for stretchable inorganic interconnects vary according to the application, the deposition method, the substrate and the required stretchability. These factors define the exact geometry of the final design, however, certain design categories can be identified. The most common design applications to be highlighted are the Origami and Kirigami based stretchable semiconductor platforms, and the serpentine/horseshoe/meandering stretchable metal structures. Variations in the design and geometry may vary depending on the intended application as shown in [Figure 10](#).

4.3.1. Origami and Kirigami Design Implementations

Following the same concept of patterning inorganic semiconductor materials in order to reach stretchability under low strain conditions, macro-scale bending and folding techniques, such as origami and kirigami, can be used to transform two-dimensional (2D) planar sheets into three-dimensional (3D) out-of-plane macro-, micro-, and nano-structures.

This structural techniques is implemented in optical, electronic, optoelectronic and micro electromechanical system (MEM) -based devices, enabling novel functionalities for applications in sensing, communications, energy storage, robotics, medicine and others [160, 161]. Previously, many cultures have used origami techniques for aesthetics and artistic purposes, generating 3D structures out of folding planar paper sheets. Kirigami is a variation of origami in which the 2D sheets are not only folded but also cut in appropriate locations to allow for more complex 3D structures. Such folding and assemblies can be achieved at a microscopic scale by means of manual arrangement of materials, using precise tools and mechanisms such as shrinking, swelling, strain mismatch, and capillary or magnetic forces, enabling the exploitation of new set of material properties [160]. Among these self-folding techniques, compressive buckling mechanisms can be used by integrating pre-patterned inorganic semiconductor sheets with pre-strained polymeric substrates. This assembly permits diverse and complex structures and out-of-plane devices with extended functionalities (Figure 10a and b) for applications in stretchable photodetectors or mechanically tunable inductors and optical devices [161-163].

A relevant property of 3D assemblies based on origami or kirigami techniques is their ability to form mobile and shape-changing structures, which can be useful in, as example, in robotic applications [160]. Additionally, extreme deformation could be desirable in order to reach large motions. Therefore, the capability of controlling the level of stretchability, from low to very high values, exhibited by the inorganic semiconductor structures is of extreme importance. Such is the case of the spiral-based architecture previously discussed, where the stretch ratio can be easily controlled by adding or removing turns from the spiral. In the implementation demonstrated by Rojas et al., a stretchability over 1300% was achieved with

a spiral structure of 200 μm in diameter and 2- μm -width arms with only 4 turns for each arm [157]. Adding more turns to the spiral design would increase the stretch ratio proportionally (for instance, one additional turn for each arm would add another $\sim 360\%$ more in the maximum stretch ratio), which allows to control the level of stretchability and thus adjust to the desired structure or level of large motion required for a specific application. **Figure 10c** shows arrays of silicon hexagon islands interconnected by spiral structures with a compliant architecture over curvilinear shapes.

4.3.2. Fractal Serpentine Structures

Most stretchable metal interconnects have a polymer and metal bilayer. This bilayer is important because metal thin films alone do not possess enough elastic restoration force to reform after a deformation [42]. The polymer backing provides the restoration force required to retake the original shape of the spring. In 2015, Hussain *et al.* showed a stretchable thermal patch using a metal polymer bilayer [164]. The polymer used in this case was lithographically defined polyimide, while the metal was electrochemically grown copper (**Figure 10d**). The device was used for thermotherapy, wherein non-stretchable islands of copper coils interconnected with stretchable lateral springs heat upon the application of voltage for pain relief. Various other studies have used copper as a metal layer on either polyimide, polyethylene naphthalate or PDMS for various applications, such as thermotherapy [165], EEG recording [166], RF interconnects [167], lighting modules [168] and thin film inductors [169]. This design has also been successfully applied using other metal thin films, such as gold [170], aluminum [171], silver [172], AgNWs [173] and ITO [174, 175]. The mechanics of bending, stretching and stress strain distribution along the length of springs have been

well studied for the meandering spring architecture [178-181]. In 2014, Rahimi *et al.* directly sewed copper wires onto a stretchable substrate in a meandering pattern. The spool was loaded with polyvinyl alcohol thread for support [182, 183].

Fractal patterns have been considered for enhancing the stretchability of basic meandering structures. A fractal is a mathematical set that can repeat itself indefinitely at any scale. Fractal or self-similar patterns have the advantage of a stretchable interconnect design because of the massive increment in stretchability achieved due to the repetitive behavior of these designs. Fractals were first demonstrated by Xu *et al.* in 2013, who used self-similar stretchable interconnects between battery nodes to form a high capacity stretchable battery [184] (**Figure 11a**). Subsequently, the mechanics and theory for fractal interconnects of various orders were reported [185-187]. Fan *et al.* demonstrated the use of fractal patterns of various types for epidermal electronics and RF applications (**Figure 11b**) [188]. The Archimedean spiral structure theorized by Lv *et al.* in 2014 [189] is a stretchable spring design that can provide higher stretchability for the same areal coverage and contour length (**Figure 11c**).

In 2015, Hussain *et al.* used metal itself as a stretchable antenna such that the resonance frequency and gain of the antenna did not shift with stretching up to 30% strain [190] (**Figure 11d**). This was achieved by patterning the metal in a meandering spring structure. When a lateral force was applied, the metal spring stretches by twisting out of plane. This deformation absorbs the work done in stretching and prevents the metal thin film from experiencing significant strain. As a result, the length and resistivity of the metal remain the same, maintaining antenna performance above the elastic limit.

5. Future Outlook and Concluding Remarks

For over four decades, electronics have been integrated in our daily life in many powerful ways. Computation, communication and entertainment have been blended as infotainment, mainly enabled through constant physical scaling of CMOS technology. CMOS electronics offer critical advantages including performance, energy efficiency, ultra-large-scale-integration density, reliability and affordability. On the other hand, state-of-the-art CMOS electronics are bulky and fixed in size, hindering their use in freeform applications. Much like the evolution of nature, technology and electronics are evolving to embrace the Internet of Everything, enabling conformal integration of electronics with living beings (e.g. humans, plants, and animals). However, body contour irregularities, asymmetry, and soft tissue challenges attempts in integration. Unlike modern rigid electronics, the natural world requires dexterous response in the form of twisting, bending, tension, and contraction. For instance, temporal volumetric changes (*viz* relaxation and contraction of muscles) is one of the most abundant phenomena in nature. Hence, to integrate with dynamic living surfaces, i.e. organs and tissues, electronics require drastic reconfiguration. In that regard, polymers offer natural shape-changing capabilities, coupling well with changing surface structures. Although polymers offer intrinsic malleability, stretchability, and optical transparency, they exhibit limited charge transport behavior and thermal instability at high temperatures. In contrast, widely used semiconducting materials such as silicon and gallium nitride, demonstrate electrical reliability and compatibility with a high thermal budget, but traditionally their bulk form renders them unreliable under even slight strain. Significant ongoing research focuses on overcoming barriers in polymeric materials by either creating conjugated heterogeneous materials, or redefining metrics from sheer performance to cost,

opening interesting applications for scientific exploration, but offers limited use in practical applications. Thus, multidisciplinary approaches seek to enhance the mechanical adaptability of traditionally reliable, conductive and semiconducting materials, while retaining their advantageous attributes.

Over the years, Prof. JA Rogers *et al.* has presented an exciting approach to material reconfiguration from a geometric standpoint allowing the hybrid integration of mechanical fractal design with classically rigid thin films (silicon, germanium, gallium nitride, etc.) enabling highly stretchable electronics [180, 187, 188]. Additional development by Prof. DH Kim *et al.* in the expansion of atomically thin and sized materials, like graphene and nanoparticles, has been influential [137, 152]. We have taken a more pragmatic approach for immediate implementation of stretchable CMOS technology enabled stretchable electronics by fusing three mechanisms: (i) fractal design enabled stretchability of thin films; (ii) use of naturally mechanically deformable (predominantly stretchable) polymeric materials for packaging; and finally (iii) pre-straining techniques. Using this combined approach, we have achieved unprecedented stretchability in thermoelectric thin films and translated these advances to a dynamically reconfigurable thermoelectric generator [unpublished results]. This technology empowers users to dynamically tune the distance of the hot and cold end of any thermoelectric generator and maximize its power efficiency by elongating that distance. These fundamental advances enable creative applications which can significantly augment the quality of our life, as demonstrated through our established stretchable smart thermal patch [164]. Flexible and stretchable electronics have been mainly dominated by applications in large area displays, energy harvesters, storage devices and implantable electronics, however progress in wearable technologies can generate immediate impact on the quality

human life. In that regard, building interactive wearable devices necessitates robust communication platforms. We demonstrate an interesting out-of-plane stretchable helical spring based metallic antenna for far field communications up to 390 meters [190]. The operation environment (*in vivo* or *in vitro*) is a critical design consideration for intimate contact freeform wearable or implantable electronics, which adds difficulties from an operational standpoint. Material and integration challenges also arise from biocompatibility requirements, affordability, stress-strain resilience, oxidation and moisture resistance, manufacturing ease, device architecture, and circuit layout design. For example, an array of non-planar vertical FinFET CMOS used under extreme bending, and repeated stretching/contraction cycles showed limited performance and caused serious electrical failure induced by mechanical deformation [191, 192]. Another aspect is geometrical configuration and process technologies, like Kirigami, which depend on specific fragmentation along an axis or a set orientation. The presence of heterogeneous materials without sufficient selectivity leads to process development and integration challenges. Therefore, innovation in both materials and processes becomes a solid opportunity for stretchable and reconfigurable electronics through the adoption of shape-changing materials.

In the future, for truly flexible systems, stretchable interconnects will be an obvious necessity. We have reservations about the use of flexible printed circuit boards or off the shelf ICs, as we believe a fundamental design change is important and this is what we target our focus on. A rigid and fixed length interconnect will go through mechanical rapture, at a bending radius of 5 mm in a flexible system, thus innovation in low-cost, high conductivity materials, with stretchable geometry is critical. To avoid permanent elongation,

interconnects must be elastic, returning to their original state after stretching. In the past, we have observed many stretchable antennae design where permanent deformation and elongation happens and broadcast frequency changes as a result. While interesting for limited usage tunable frequency communication, they are not suitable for continuous fixed-frequency communication. Although fractal designs can enable stretching in normally rigid thin film materials, the gap creation and its mitigation are important areas to focus on. We have overcome this challenge by introducing robotic reconfiguration, a new area we fondly term, “*Robonics*”, a combination of freeform electronics and robotics. Integrating energy harvesting or storage devices using advanced functional reconfigurable materials introduces advantages, *e.g.* piezoelectric materials capable of harvesting energy when subject to mechanical deformation.

We began our discussion with the rise of electronics and evolution of modern computing, from the bulky desktop computers and trendy laptops to today’s sleek tablets and smart phones, modern revolutionary technologies are making their predecessors nearly obsolete. While we easily predict the enhancements in performance, functionality, energy management and lower costs, the convenience driven change in form factor may slip our minds. Imagine a future where a 5.5” smart phone can be stretched to a 55” television. While it may sound impractical, we have taken first steps toward this vision, showing a 10× stretching in mono-crystalline silicon. As silicon (100) is used in fabricating nearly ninety percent of the electronics on the market, and is stubbornly rigid and brittle, our efforts in stretchable and flexible silicon embolden our vision. Our demonstration of a hexagonal platform with extreme stretching ^[157], sequential contracting and potential reconfiguration lends itself to important applications in stretchable electronics. For example, lifetime heart

implants capable of changing size and shape could replace repeated surgery as children grow. While still conceptual, the opportunity exists using the spiral geometry as we can control their number of turns and can activate each of them as needed, allowing variability of size and control of the overall architecture. Such reconfiguration offers amazingly exciting opportunities from both fundamental science and engineering innovation perspectives. In addition to innovation in new materials, we have to opportunistically use existing materials, processes, and devices to bring tomorrow to today. This vision drives us more towards a CMOS centered technology embodying heterogeneous materials and hybrid processes for stretchable and reconfigurable electronics. We see a future where freeform (physical flexibility, stretching and reconfiguration) will add new dimension to existing performance, density, energy efficiency, reliability and affordability. Technology which changes our life is not always an optimal solution, it is a compromise and a multi-disciplinary effort that is often ignored in the academic publication world. In order to realize these technologies, we hope to see more pragmatic discussion and development focusing on long ranging impacts of stretchable and reconfigurable electronics to implement the Internet of Everything. To achieve the Internet of Everything, design focus must capitalize on natural phenomena: irregularity, tenderness, asymmetry in living beings, and the presence of naturally insulating, conducting and more importantly semiconducting rigid materials. An ever increasing focus on transforming rigid materials into flexible and reconfigurable forms broadens horizon of electronics, and offers the potential to augment the quality of life.

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Figure 3. Preparation Processes of Hybrid Elastomers. (a) Manufacturing process of SWNT film, SWNT elastic conductor, and SWNT paste for a stretchable active matrix OTFTs. From [66]. Reprinted with permission from AAAS. (b) AgNWs composite fibers prepared through a dip-coating process. Reprinted with permission from [50]. Copyright 2015 American Chemical Society (c) CNT/Graphene/PDMS hybrid mixed composite. Schematic displays the interaction between CNTs and graphene. Reprinted with permission from [83].

Copyright 2016 American Chemical Society. **(d)** Formation of MeTro/GO hybrid hydrogel, where schematic on the left shows the effect of twisting on the structure of MeTro/Go elastomer. Reprinted under the Creative Commons Attribution License (CC BY) from [\[41\]](#).

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