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#### **SURVEY PAPER**

#### New materials and advances in making electronic skin for interactive robots

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Flexible electronics has huge potential to bring revolution in robotics and prosthetics as well as to bring about the next big evolution in electronics industry. In robotics and related applications, it is expected to revolutionise the way with which machines interact with humans, real-world objects and the environment. For example, the conformable electronic or tactile skin on robot's body, enabled by advances in flexible electronics, will allow safe robotic interaction during physical contact of robot with various objects. Developing a conformable, bendable and stretchable electronic system requires distributing electronics over large non-planar surfaces and movable components. The current research focus in this direction is marked by the use of novel materials or by the smart engineering of the traditional materials to develop new sensors, electronics on substrates that can be wrapped around curved surfaces. Attempts are being made to achieve flexibility/stretchability in e-skin while retaining a reliable operation. This review provides insight into various materials that have been used in the development of flexible electronics primarily for e-skin applications.

Keywords: electronic skin; novel materials; tactile sensing; robotics

#### 1. Introduction

The rapid advancement in technology in the last few decades has now enabled development of robots which has long been a mere concept in science fiction movies. From its primitive stage as controlled industrial tool operating human restricted environment, robots have evolved into autonomous and self-adapting systems to variant situations. Furthermore, robots such as humanoids are expected to be involved in day to day human interaction, therefore it is also critical to build a safer system which can interact with human. One possible approach of building such system is by inducing the sense of touch to robots.[1]

Among various human senses, sense of touch plays a crucial role in the way in which we perceive our environment. For instance, information such as surface roughness, temperature and size which are critical for object discrimination and manipulation can only be determined by the sense of touch.[2] Inspired from human skin, the development of artificial skin (also referred to as synthetic skin or electronic skin (e-skin)) has become an area of immense interest to scientists. The primary function of e-skin is to provide tactile information which could be used to evaluate aforementioned parameters for the object handling. In addition, tactile sensors can also provide information on surface compliance, hardness of object and electrical conductivities. [3–5] Other possible functionality which could be embraced by e-skin are chemical, temperature and biological sensors. Furthermore, development of self-healing materials are currently under investigation.[6]

In addition to development of sensors, integration of sensors over a large area is also a critical requirement for the development of efficient e-skin. Furthermore, distribution of sensors over non-uniform (or curvy) surface would provide an improved performance due to higher distribution of sensors.[7] In addition to robotics, e-skin can also have an influence in applications such as personal health care monitoring,[8] wearable technology,[9] artificial intelligence and medical prosthetics.[10]

This paper presents an overview of various material used in the development of e-skin which is critical components in tactile sensing. However, unlike in previous review in tactile sensing, we have primarily focused on the materials used for the development of e-skin sensors.[2,11]

#### 2. Tactile sensing for humanoids

Tactile sensing for robots has been studied since 1980s. In the context of humanoids, tactile sensing is primarily used

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in the effort to replicate the human sense of touch through a smart interplay of hardware and software. It is one of the fundamental sources of information required for accurate perception, and is essential to any tasks requiring object manipulation, gripper dexterity or interaction with an unknown, cluttered environment. Tactile sensing can greatly assist vision systems by providing information unobtainable solely from image data, such as weight, surface texture or stiffness. A simple example illustrating the importance of this sense is the difficulty of tying shoe laces together with numbed fingers; such a task is extremely difficult; however, with the sense of touch restored it becomes trivial (at least for a robotic system that has learned how to tie shoes).

Development of flexible and stretchable electronic skin can inspire new functionality and precise tactile information. The electronic skin depending on the material and sensor architecture can be utilised to measure normal and sheer forces, vibration temperature and nociception.[12] Furthermore, e-skin also provides a pathway for a safer human-robot interaction. Through tactile interaction, various touch or contact modalities may be carried out; a robot may be patted, slapped, punched, or tickled, with each action representative of a separate communicative intent. For any robotic system that is to work closely with humans, evaluation and classification of these touch modalities is vital.[13] In other words, humanoids should understand, just as humans do, that a slap is a form of negative feedback that a pat is one of encouragement and so on.[14] Moreover, having flexible, stretchable tactile sensors over whole body of humanoids are particularly important in applications such as disabled and aged care, nursing and caring for patients with mild mental impairment, where a significant amount of communication is non-verbal interaction.[15]

Furthermore, touch information is beneficial for a natural handling of a robots motion by users. For instance, users can push the robot away in an arbitrary direction to place it accordingly.

Tactile sensors can be also classified based on its transduction mechanisms. Frequently used transduction mechanisms include capacitive, piezoresitive, piezoelectric, triboelectric, ultrasonic, optical and magnetic. The detailed study of some of these mechanisms are available in [12].

Over the years, there has been a paradigm shift in the development of flexible and stretchable e-skin. Early works on development of flexible e-skin involved the use of flexible PCB or PCB on flexible substrate onto which the electronic components were mounted. Examples of such work includes development of triangular [16] and hexagonal [17] tactile skin patches. Kim et al. [18] developed such a skin using silicon micro-machining and packing technology (on a flexible substrate), allowing for the detection of normal and shear forces at high resolution. This skin was shown to be able to effectively measure normal force, hardness, slip and touch. Such a sensor is ideal for touch classification since movements such as tapping and rubbing can be easily differentiated via the applied shear force. Restricting measurement to only force allows for a higher resolution, however it limits the ability to collect vibro-tactile data. RI-MAN [19] is one of the few humanoid robots capable of interacting through whole-body contact, and is able to perform complex movements such as lifting a human with its arms. Semiconductor pressure sensors were placed in multiple sections of the robot body, providing tactile feedback on the position and orientation of the human subject. However, poor bending radii limited their use in applications such as finger tips of robotic hands which require larger bending radii.

In recent years, the trend in development of e-skin has shifted to development of flexible and stretchable electronics. Nevertheless, the development of flexible and stretchable electronics has been impeded by many challenges including those posed by today's electronic systems, which are developed on rigid and non-planar substrates. Development of flexible and stretchable electronic systems requires novel and cost-effective fabrication techniques, new materials that lead to innovative devices and structural designs that can withstand large strain or deformity during their use. In addition to technological aspects of the sensory device, the performance of robotic system equipped with e-skin also depends on the appropriate processing and learning methods that interpret information contained in tactile data. For instance, dexterous object manipulation with an anthropomorphic hand requires flexible electronic skin which can provide high enough spatial tactile resolution. Flexible and bendable robotic skin provides the robotic hand with the ability to accurately detect and accordingly to learn the physical properties of in-hand object for dexterous in-hand object manipulation.

The haptically accessible object characteristics can be divided into two general classes: geometric and material properties. The geometric properties can be recognised by the object size and shape and the related work can be found in [20-22]. The object material can be characterised and differentiated based on surface texture, stiffness and thermal quality obtained through tactile sensing. For instance, in order to classify cotton, linen, silk and denim fabrics, Song et al. designed a mechanism to generate the relative motion at a certain speed between the PVDF film and surface of the perceived fabric. In this study, neural network and K-means clustering algorithms were used for fabric surface texture recognition.[23] Five textiles were explored and discriminated from each other via k-nearest neighbour (K-NN) using an active sliding touch strategy and an array of microelectromechanical systems (MEMS) in the distal phalanx of a robotic finger.[24] Jamali et al. fabricated a biologically inspired artificial finger composed of silicon within which were two PVDF pressure sensors and two strain gauges. The finger was mounted on a robotic gripper and was scraped over eight materials. The Majority voting learning method was employed to find the optimal



Figure 1. Robots equipped with e-skin. (a) NAO Humanoid Robot with multi-modal artificial skin, at the Institute for Cognitive Systems, TUM- Germany; (b) the Shadow Robotic Hand with BioTac Sensors on fingertips at the Shadow Robot Company UK.

technique for the texture recognition problem.[25] Kaboli et al. proposed a set of biologically inspired tactile feature descriptors to classify and categorize objects through their texture and weight, respectively [14]. In this respect, the NAO humanoid robot with multi-modal artificial skin on the arms was employed. The NAO humanoid equipped multi-modal artificial skin is shown in Figure 1(a). In [15], for the first time, an anthropomorphic robotic hand called Shadow Hand with artificial skin on the fingertips, shown in Figure 1(b), discriminated 20 different in-hand objects with different shapes via the surface texture properties.

## **3.** Materials for the development of stretchable electronics

The ability to stretch, flex and self-heal on occurrence of damage are some of the defining features of human skin. In addition, its extraordinary sensing capability to detect a broad range of force further signifies its importance to human. It is critical to incorporate some of these features into e-skin to achieve better performance from robots that could match or rival the performance of humans. Therefore, the choice of materials for the development of electronic skin is critical as they greatly influence both the mechanical and electrical performance of the device. Stretchable electronics are realised via: (a) synthesis of novel materials such as composites of soft materials with conductive fillers (b) Smart structural engineering and designs such as serpentinelike structures for interconnects or wires. Furthermore, flexible nature of materials could also facilitate towards low cost and large area fabrication such as roll-to-roll production. An overview of various material used in the development of stretchable electronics will be presented in the forthcoming section.

#### 3.1. Substrates

Silicon has an unprecedented impact on the electronics industry over the last several decades and its wafer has become the natural choice as a substrate for new developments in modern electronics. However, the rigid and brittle nature of Si wafer limits its use in the development of flexible electronics applications. Among various polymers, elastomers like PDMS have received a significant attention due to the biocompatibility, chemical inertness and mechanical strength (Young's Modulus of 1.8MPa).[26-28] An apparent advantage of elastomer for e-skin application is its conformability to uneven surface, thus aiding distribution of sensors. Rogers's group demonstrated that higher strain could be accommodated by building devices on top of islands moulded on top of PDMS. In this scenario the islands were capable of withstanding a strain of 452% between the trenches presented within the island while the strain at top and bottom surface of the island were 0.32 and 0.36%, respectively.[29] In another study, Yamada et al. demonstrated carbon nanotube strain sensor with PDMS as a substrate. The reported device was capable of accommodating strain of up to 280%.

Other polymers such as Ecoflex,[30,31] polyimide (PI), [32,33] polyurethane and poly(ethylene naphthalate) (PEN) [34] have been investigated as suitable substrate for flexible electronics applications. A significant advantage of Ecoflex in comparison with the other polymer is its biodegradability.[30]

#### 3.2. Dielectric

Dielectric materials are one of the most critical components in the development of flexible electronics devices. Some of the key criterions expected of dielectric materials are high capacitance and low temperature processability. High capacitance layers are preferable for a low voltage or highperformance operation. PDMS has been one of the versatile materials in terms of its applications for the development of flexible and stretchable electronics, in addition to its use as a substrate, PDMS has also been exploited as a dielectric material. Furthermore, it has also been demonstrated that micro structuring of PDMS film results in an improved device sensitivity and device performance for pressure sensing applications.[8,35] Microstructures on PDMS allow it to elastically deform on application of an external force, thereby storing and releasing the energy eventually leading to the reduction of the viscoelastic creep. Besides PDMS, other polymers such as polyimide,[34] co-polymers such as P(VDF-TrFE) [36,37] has also been used as dielectric for pressure sensing applications. Other materials that have been investigated as dielectric material for flexible electronics applications include polymer composites comprising of nanofillers, high-K dielectric materials and liquid ion gels.[38] Some of the high-K nanomaterials used to develop hybrid dielectric materials includes TiO2 [39] and BaTiO3.[40] In addition to other high-K dielectric composite, high-K materials like aluminium oxide (Al2O3),[32] tantalum oxide (Ta2O5)/SiO2 [41] have also been used as gate dielectric. Ion gels, also referred to as gel electrolyte comprises of an ionic liquid and a co-block polymer.[38] Ion gel provides a very high capacitance as a result of nanometer thick double layer formation at the electrodeelectrolyte interface and it proves to be an ideal material for gate dielectrics. Furthermore, faster response time, high frequency operation (10 kHz) and solution process compatibility makes it a suitable dielectric material for flexible electronics applications.[42] Sun et al. [43] reported a development of coplanar gate graphene field effect transistor matrix comprising ion gel as a gate dielectric for pressure sensing applications.

#### 3.3. Active materials

#### 3.3.1. Nanowires

Nanowires (NWs) of semiconducting materials are one of actively researched materials for the development of flexible and stretchable electronics due to their excellent electrical and mechanical properties.[44–46] NWs of inorganic materials are attractive choice for realising electronics for robotic skin applications. In particular, semiconducting NWs possess interesting electrical, optical, mechanical and electrochemical properties, which would be ideal for applications such as nanoelectronics, sensors, optoelectronics and photovoltaics applications. Some of widely used NWs includes zinc oxide (ZnO),[47] germanium (Ge),[33] Gallium arsenide (GaAs), InAs [48] and Silicon (Si). [49,50] Though a significant progress has been made on the synthesis of NWs via top-down and bottom-up approach, the higher cost associated with synthesis of NWs and difficulty in obtaining a highly aligned uniform NWs limits its potential use in large area electronic applications. In a recent work, Javey group have demonstrated fabrication of pressure sensors on a polyimide substrate suitable for large area electronics applications including electronic skin. The developed pressure sensor was based on Ge/Si core-shell NW FET (Field Effect Transistor) which was grounded via a pressuresensitive rubber (PSR).[33] Application of pressure causes a change in conductance of PSR thus affecting FET characteristics in a manner similar to POSFET (Piezoelectric Oxide Semiconducting FET) tactile sensing devices,[51,52] which we developed in past and the organic FET-based pressure sensors reported by Someya group.[34] Among the compound semiconductors, ZnO NWs have been shown to be tactile sensing element based in piezotronic transduction mechanisms.[53] As grown ZnO films have been used in fabrication of large area self-powered tactile imaging circuit. This brings an opportunity to directly integrate material synthesis, device fabrication and mechanical actuation. As against conventional vertical wrap gated FETs,[47] ZnO piezotronic transistor consisting of metal-semiconductormetal junctions which utilises polarisation of immobile ions for device operation has been demonstrated. The channel conductivity of ZnO piezotronic transistor was modulated by externally applied stress over the metal surface. The externally applied strain causes polarisation of ZnO, which affects the transport characteristics. Hence, the transport characteristics are affected by externally applied strain, which effects the polarisation in the ZnO NWs. The reported taxel density of the strain gated piezotronic array is  $8464 \text{ cm}^{-2}$ , which is 35 times higher than that of the mechanoreceptors in human's fingertip. Also, the pressure sensitivity values match with the human skin, i.e. few kPa to 30 kPa. These merits clearly demonstrate the reduced gap between the human skin and the artificial e-skin. In another study, a highly sensitive pressure sensor was developed by incorporating tissues impregnated in gold NWs between two PDMS substrates- of which the bottom substrate consisted of interdigitated array of electrodes. The change in pressure is detected by monitoring the change in resistance of the device. In addition to pressure, this device was also capable of differentiating between various mechanical stimuli such as bending, torsional, pressing forces and acoustic vibration. In addition, the device is reported to be scalable, in which case the approach is ideal for large area fabrication. However, additional sensing/functional capabilities such as temperature

sensing, texture recognition, distributed heating and signal processing need to be added in these approaches to make them perform at par or better than human skin. This is very well possible using Si NWs based approach for artificial skin in tandem with various sensors realised with inorganic NWs. Si nanoribbons-based transduction mechanisms for e-skin has been demonstrated to sense light and temperature. An e-skin with all these sensors could find powerful and interesting applications in robotics. However, the transfer related issues delay utilisation of the full potential of the elementary and compound semiconductor NWs. Current transfer printing processes need to be scaled up for large area printing. Figure 3 depicts fabrication steps for top-down synthesis of silicon microwire and the subsequent transfer printing process. Development of new manufacturefriendly transfer process certainly helps to benefit more from semiconducting NWs. One such initiative is the printing of electronic layers from NWs, which we are investigating through PRINTSKIN project.[54]

#### 3.3.2. Carbon based materials

Graphene and carbon nanotubes (CNT) are two extensively studied carbon allotropes owing to their fascinating material properties. Their intrinsic material properties such as near ballistic transport [55,56] and extraordinary mechanical properties [57,58] offer a new perspectives for the development of sensing technologies over ultra-thin substrates. As with any novel materials, the potential of these materials for e-skin and related approaches relies heavily on the capability to develop a reliable fabrication methods with low cost and scalability. Solution process techniques such as spin-coating,[59] spray-coating [60] and ink jet printing [61,62] are some of the methods which could boost the potentials of these materials in the development of large-scale devices. Mechanisms of the above techniques are explained in detail in a review by Khan et al. [63].

Lipomi et al. [64] reported the development of transparent and stretchable electronic pressure sensor capable of detecting a pressure around 50 kPa. The sensor comprised of Ecoflex layer sandwiched between two CNT thin films spray-coated on top of a PDMS substrate. In addition to pressure sensing, the device was also capable of detecting strain. The application of strain or pressure causes a change in capacitance, which was used as transduction mechanism to sense the registered pressure or strain. Simple fabrication process, physical robustness and mechanical compliance of this sensor are some of the key features that can be exploited for large area electronics applications. In another study, by developing a thin film of highly aligned single wall CNT (SWCNT) on a PDMS substrate, Yamada et al. [65] developed a strain sensor capable of withstanding a strain as high as 280%. Furthermore, the device also exhibited high durability, low creep time and faster response time. In addition, electrical response of device remained unchanged even under a higher strain after a prolonged cyclic test. The performance of device was merely limited by the PDMS substrates, which began to rupture under a repeated cycle at 200% strain. Such reported sensors are suitable for human motion detection sensors and for wearable electronics applications. Additionally, CNT-based devices on unconventional substrates have also been demonstrated for applications such as flexible CNT transistors,[66] bendable vapour sensors [67] and flexible pH sensors.[68,69] Development of such devices could pave way for the development robotic skeleton system with integration of sensors for various applications.

Besides CNT, graphene is another potential candidate for the development of e-skin components for robotics and similar applications. Since the isolation of graphene in 2004, [55], a great progress has been made in the synthesis of large area of graphene. Wafer scale growth of high quality graphene is possible via chemical vapour deposition (CVD) on a metal surface [70-72] and epitaxial growth of graphene on SiC (high temperature and expensive process).[73] Methods such as chemical exfoliation of graphite are other promising routes for large-scale production of graphene for large area electronics applications.[74] The development of graphene-based devices for stretchable and flexible electronics requires transfer printing of graphene to various substrates. The transfer printing often leads to degradation of graphene due to the formation of cracks or due to residual remains of the support layer used during the transfer printing process.[75,76] However, recent development on transfer printing has led to crack and residuefree transfer printing process.[77] Similar to CNTs, solution processing techniques such as inkjet printing, [78] spray coating [79] are another viable solution for the realisation of graphene-based devices. However, in comparison with CVD graphene, solution processed graphene exhibit poor uniformity and higher sheet resistance. Transfer free synthesis of graphene is another option for realisation of graphenebased device on flexible substrates. Graphene is an excellent material for the development of thin film transistors (TFT) for flexible electronics applications.[80-82] It exhibits both metallic and semiconducting properties, which has been utilised to develop all graphene based TFTs. For example, Ho-Cho's group developed all graphene-based coplanar graphene FET (GFET) with an ion gel as gate dielectric.[81] The GFET exhibited high mobility, low voltage operation and high on-current. Under strain (up to 2.8%) a 20 % change in the carrier mobility of the device was observed; furthermore, no prominent change in the device performance was observed under ambient conditions. In a different study, based on the same co-planar gate geometry, the same group developed a low power pressure sensor for e-skin applications. The device had a high sensitivity of 0.12 kPa-1, low operation voltage and good mechanical stability.[43] These features are very attractive for e-skin in robotics, where fast, reliable and repeatable response is



Figure 2. Large area graphene for flexible device applications. (a,b) Transfer printing of roll-to-roll fabricated 30 inch graphene and the resistive graphene based touch screen film using flexible polyethylene terephthalate (PET) substrates [87].(c) Schematic illustration and the operation of the broadband optical modulators based on graphene supercapacitors [121]. Application of bias voltage through the electrolyte medium dopes the graphene layers and yields a transmittance change with the blocking of interband transitions in graphene. (d) Graphene based flexible electrochromic devices [88].

much desired.[2] Other graphene-based solutions, which can also be used in robotic systems, are flexible and transparent strain sensors, [83] flexible supercapacitors [84] and gas sensors on bendable and soft substrates.[85,86] In that sense, current focus of the graphene research is the fabrication of large area graphene electrodes on flexible substrates for touch sensor and smart window applications. Figure 2(a)and (b) show the transfer printing of roll-to-roll fabricated 30 inch graphene and touch sensor panel developed using graphene on flexible polyethylene terephthalate (PET) substrates [87]. Figure 2(c) shows the schematic illustration and the operation mechanism of the broadband optical modulators based on graphene supercapacitors [121]. Application of bias voltage through the electrolyte medium dopes the graphene layers and shifts the Fermi level. This causes the blocking of interband transitions in graphene which makes the material more transparent. Figure 2(d) shows the graphene based flexible electrochromic devices [88]. Multilayer graphene changes its colour with the intercalation of ions through the graphene layers. At 5 V device allows to see the University of Glasgow logo placed beneath it. Device operation is stable under mechanical stress.

Given these developments, the integration of graphene sheets onto flexible, ultra-thin and soft substrates could find variety of uses in robotic skin ranging from motion sensing to display applications. For example, the usage of large area graphene sheets as flexible and transparent electrodes [87] in the robotic skin would yield to advanced sensing of many environmental parameters due to high carrier mobility and high surface coverage. Alternately, usage of graphene-based flexible electrochromic devices [88] and/or graphene/nanotube-based smart windows [89] could provide skin like display panels over the limbs of a humanoid to show the information about the current status of the system.

#### 3.3.3. Organic materials

Organic semiconductors fuelled the initial developments in the field of stretchable and flexible electronics. Though these materials exhibit a poor mobility in comparison with inorganic semiconductors, the low cost and large area fabrication compatibility are some of their advantages. Organic materials have tremendous prospective applications for electronic skin applications. Some of the widely used conductive polymers include poly (3,4-ethylene dioxythiophene):polystyrene sulfonate (PEDOT:PSS), poly (3-hexylthiophene 2,5-diyl) (P3HT), polypyrrole and polyaniline (PANI). These conductive polymers can be used as conductive fillers in the composite.[12] Organic semiconductors are widely used materials in the development of flexible electronics. Some of the organic semiconductors used in the development of e-skin includes pentacene and rubrene.[34,35]



Figure 3. Schematic of fabrication steps of silicon wires by topdown approach and the subsequent transfer printing of the wire to a target substrate using PDMS as supporting layer; (a)–(b) silicon microstructure were obtained by lithographic patterning and deep ion reactive etching of SOI wafer; (d)–(e) silicon microwire transferred using a plasma exposed PDMS; (f)–(h) silicon microwires transferred to the target substrate and finally the PDMS was removed.[45]

#### 3.3.4. Stretchable polymer composite

Polymers are an interesting class of materials for the development of flexible and stretchable electronics owing to their excellent mechanical properties. Nevertheless, the poor electrical conductivity of these materials limits their use to substrate and dielectric applications during the development of flexible electronic. Introduction of conductive fillers into the polymers results in a composite with high electrical conductivity and mechanical stretchability.[90-92] Though polymer composites have long been investigated, its potential application has been limited by high filler concentration which has a negative effect on the mechanical properties of the composites.[93] Issues such as high filler concentration can be obviated by the introduction of nanomaterials. Unlike traditional conductive fillers, the use of nanomaterials as fillers could enable the composites to acquire the desired property at a lower filler concentration.[94,95] Some of the widely used nanofillers are graphite nanopallets, NWs, carbon nanotubes (CNT) and graphene. The high aspect ratios of nanofillers such as CNT and graphite flakes are the key reasons behind the low percolation threshold of these materials. Percolation threshold is the minimum volume fraction of the conductive filler required for the transition of the polymer from its insulation to conduction phase.[12]

The transition of the polymer from its insulating to conductive phase occurs when the concentration of the fillers exceed the percolation threshold, resulting in a formation of conductive network within the polymer matrix. A lower percolation threshold is preferable to retain the elasticity of polymer. The percolation threshold can be influenced by various factors such as type of polymer matrix, size, shape, aspect ratio and surface condition of fillers.[96] In addition to aforementioned parameters, uniform dispersion of fillers within the polymer also plays a crucial role in achieving a lower percolation threshold.[97] Dispersion of fillers within the polymer can be achieved by various techniques such as sonication, [98] ball milling, [99] mechanical stirring, sheer mixing [100] and surfactant-assisted process. Carbon black an amorphous form of carbon is an attractive candidate for the development of conductive fillers have been used in the development of flexible tensile stress and pressure sensors.[101] Among the list of other filler materials, CNT is another attractive candidate as a nanofiller.[94, 95] The high aspect ratio of CNT often leads to a lower percolation threshold. The realisation of nanocomposites and devices is greatly influenced by patterning technologies. Among the various patterning techniques of composites, the most common technique is moulding. As depicted in Figure 4(a), a master mould made of SU-8 or other UV curable material is fabricated with desired structures patterned by standard photolithography. The prepared nanocomposite is then poured onto the mould and cured. Following curing of composite, the polymer substrate is poured on top of the patterned conductive composite on the mould. Finally, the bilayer film consisting of polymer substrate/conductive composite is peeled from the mould resulting in a patterned structure of the composite on the substrate.[102,103] Another popular patterning technology is the micro-contact printing which relies on the PDMS mould made by casting or machining and transfers the conductive composites onto polymer substrate shown in Figure 4(b).[104]

#### 3.4. Smart structural engineering

Traditionally, electronics has been developed via use of inorganic materials and metals. Development of stretchable electronics utilising these material is highly favourable due to their superior electronic performance and mature fabrication technology. However, the use of these materials is limited due to their rigid and brittle nature. Dahiya et al. [105] reported development of piezoelectric oxide semiconductor field effect transistors (POSFET) for robotic tactile applications. The device demonstrated a sensitivity of 102.4 mV/N. Though the device exhibited a good sensitivity, its use in electronics skin application is limited due to the rigidness of the POSFET. Such hurdle in the development of flexible electronics through the use of intrinsically brittle material can be overcome by the smart structural engineering of the materials, to accommodate the strain caused due to



Figure 4. (a) Moulding process for patterning CNTs-PDMS composite.[88] (b) Micro-contact printing for patterning CNT-PDMS composite.[104]

flexing of the materials. Some of the widely used structural engineering techniques includes: (1) Thinning down of Si wafer (2) Buckling; (3) Use of stretchable interconnects to connect rigid islands.[106]

#### 3.4.1. Ultra-thin Silicon chips

The organic semiconductor-based analogue and digital electronics is not sufficient to meet many challenges, especially those related to high performance requirements and stabilities. They are severely unstable to design analogue circuit and sensor blocks such as comparators, amplifiers and ADCs.[107] This is mainly due to low charge carrier mobility of organic semiconductors, which results in devices that are much slower than their inorganic counterparts. To overcome these challenges, new forms of high mobility material such as single crystal Si nanowires and ultra-thin chips have been investigated. Although very promising, the Si micro-/nanoscale structures based approach is still at infancy. On the other hand, ultra-thin flexible chips are promising as they enable compact electronics and are bendable.

Si chips are traditionally built on wafers whose thicknesses are in the range of 100 micrometres. These wafers are intrinsically brittle, thus limiting their use in the development of flexible electronics. Flexibility can be induced into Si wafer if it is thinned below 50  $\mu$ m, in the range of 20–50  $\mu$ m. In addition at 10  $\mu$ m range, the Si exhibited a transparent nature, therefore enabling its usage in displays applications.[108] These ultra-thin flexible Si chips can be transferred onto a polymeric foil to form system in foil (SiF) devices for electronic skin applications.[109] Thinning of Si chips are generally achieved either by physical or chemical methods.

Among the physical methods, back grinding of wafer is the most popular method for thinning of wafer using a grinder wheel. Traditionally, the removal rate for back grinding ranges from  $0.1 - 100 \mu m/min.[110,111]$  The back grinding of the sample causes sub surface damage and crack at the edges. The thinned wafers are transferred using a carrier wafer, following which the thin membrane is eventually removed.[112] In addition, thin Si-based devices and nanomembranes can also be achieved by chemical etching of SOI (Si on Insulator) wafer. Chemical etching of Si can be achieved either via both dry and wet etching process. The thinned Si is removed from SOI wafers by etching the underlying oxide. Some of the widely used wet etchants of Si are ethylenediamine prrocatechol (EDP), potassium hydroxide (KOH), tetramethylammonium hydroxide (TMAH). Wet etching of samples lead to undercutting,[113] which could be evaded by using dry etching process. Common dry etching techniques include: (1) Plasma systems; (2) Ion etching; (3) Reactive ion etching. High cost of SOI wafer is another limiting factor. Various alternates for SOI wafers have been proposed. Some of the techniques include Dicing Before Grinding (DBG),[114] thinning of wafer by a combination of selective wet etching and back grinding process- The devices are fabricated on top of epitaxial grown Si. Other available techniques for thinning of chip includes Chip film, Hyperion and Taiko [108,115,116].

Despite progress and achievement of the ultra-thin Si chips in improving of the bendable electronics, the conventional BSIM (Berkeley Short-channel IGFET Model) models fail to predict the behaviour of such devices since they are appropriate for rigid and planar structures. These models need to characterise and capture the effects related to uniaxial, biaxial and shear stress, which is important from circuit design aspect as well as various bendable electronics applications.



Figure 5. Fabrication process flow for engineering 3D buckled structure. (a)–(g) Process steps in the formation and 3D buckled semiconductor nanoribbons and process of incorporating it within the PDMS. (h) Response of the semiconductor nanoribbon under application of force. [118].



Figure 6. Percolation path of gold film on a PDMS substrate (a) SEM micrograph of gold film on a PDMS substrate at different strain from 0–20%. The random cracks observed at 0% strain is due to mismatch in thermal expansion coefficient PDMS and gold film occurred during the cooling process following the deposition of gold film. (b) Schematic of conduction percolation pathway on gold film at 0% strain and 20% strain.[120]

#### 3.4.2. Formation of wavy patterns

Buckling is another technique that enables stretchability of intrinsically inelastic material. It is achieved by depositing a thin film of thin inelastic material on top of a pre-strained elastomer. Wavy patterns are formed on relaxation. Uniaxial strain of elastomer results in a linear waveform while a biaxial strain results in 2D herringbone structure.[117] In a study by Roger's group, a pop-up structure consisting of Si and GaAs nanoribbons were fabricated and these structures were capable of withstanding a stretchability and compressibility of 100 and 25%, respectively. [118]. The fabrication process flow of this pop-up structure is depicted by Figure 5. A significant advantage of buckling technique is that it enables the use of inorganic semiconductor material. Buckling has been demonstrated in various materials ranging from metallic, semiconducting and CNT thin films. Lipomi et al. [64], demonstrated a transparent pressure and strain sensor based on the buckling mechanisms. The developed sensors withstood a strain of 150% along with high conductivity. Other similar approaches to attain buckling include structures such as serpentine and coiled spring etc. These pop-up structures can also be exploited as stretchable interconnects connecting two rigid sections of the circuits.[119]

#### 3.4.3. Formation of micro-crack patterns

Formation of intentional micro-cracks is one approach that could be used in the development of stretchable electronics. This is achieved by depositing a thin metal film on top of an elastomeric substrate. Continuous loading and unloading causes the formation of cracks on the metallic film, the formation of percolation path by the micro-cracks played a critical role in the conductivity of the film. Graz et al. [120] demonstrated the formation of micro-crack on a thin gold film deposited on top of PDMS substrate. The film exhibited an excellent robustness under a strain of 20% for over 250,000 cycles. Figure 6(a) shows the SEM micrographs of micro-cracks formed in on the gold film at different strain while Figure 6(b) shows a schematic of percolation conduction path at different strain.

#### 3.5. Summary

Flexibility and stretchability will be the key criterions of future electronic skin. As described above, this could be achieved either via smart structural engineering and use of novel materials. In addition to mechanical robustness, the choice of the material is also influenced by the application. For instance, PDMS could be used as both a substrate and dielectric based on applications.

The use of smart structural engineering techniques would enable the use of well-established silicon technology to develop flexible and stretchable e-skin sensors. Furthermore, sensors developed via these technique would enable better system integration thus aiding easy integration with circuits of data collection, signal condition and processing of the received data.

Novel materials such as nanowires, CNT and graphene exhibit excellent mechanical and electrical properties which are critical parameters for the development of e-skin sensors. Nevertheless, the use of these novel materials is limited by the higher fabrication cost and yet to be optimised transfer printing that would suit large-scale production of devices with similar characteristics. In addition to passive sensors, developed via various transduction mechanisms, development of transistors using the novel material are highly desirable as it would enable the development of active circuit matrix for large-scale sensing with low power consumption. It will also enable easier readout circuit and individual access to devices. Development of FET using the novel material is challenging due to various aspects. Graphene for instance is a zero bandgap material, therefore GFET often exhibit a high off-current leading high power consumption in its off state.

Organic materials are also desirable for the development of flexible components for e-skin due to low cost. Someya's group has pioneered the development of organic FET for pressure sensor applications and have developed pressure sensors suitable for robotic fingertip.[34]

#### 4. Conclusions

The development of flexible and stretchable sensors for e-skin applications has seen an unprecedented growth in recent years. This has to be attributed to development of novel material and engineering which has enabled innovative devices. Flexible and stretchable e-skin would have significant impact on the tactile sensing capability of humanoids, therefore will have a critical component of future humanoids.

Despite several reported progress in development of flexible pressure and strain sensors for e-skin application, there are still several significant hurdles that need to be addressed to enable mass production. This includes low-cost fabrication process with higher device yield with similar characteristics. Other factors such as lower power consumption, device sensitivity, device stability after repeated operation, response time and operation bandwidth are also critical factors. More importantly, device integration with system is crucial for the true success of e-skin for tactile sensing of humanoids. This would require development of an electronic interface consisting components for digitalisation of signal, signal conditioning, data processing and transmission of data. Furthermore, the performance of robotic system equipped with e-skin will also heavily be governed by the software algorithms processing and learning methods to distinguish between different tactile data. In addition to tactile sensing, other features like self-healing, chemical and biological sensing can also benefit e-skin. Recent developments have led to e-skin sensors exceeding sensitivity of human skins in terms of detection of human skin. Although there are many issues yet to be addressed, the progress in the development trend in e-skin suggest that humanoid equipped with flexible and stretchable will be possible in near future.

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