Nanomaterials in Wearable Electronics: Fabrication Techniques, Challenges and Prospects

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ABSTRACT:

Wearable electronics incorporating smart nanomaterials with excellent electrical properties and great flexibility have revolutionized wearable applications like healthcare surveillance, human−machine interfaces besides developing electronic skins mimicking real skin. Despite the advancements such as ability to notify changes in advance, monitoring various properties of active nano-materials accurately and conglomerating various nano-materials to fabricate tailor made products without compromising functioning, electronic skin akin to real skin still have limitations. Thus, here an overall assessment of the skin-oriented electronics is provided by reviewing various works in this field. Various approaches in synthetization of soft and flexible nano-materials, to be employed as components for the production of flexible sensing devices, optoelectronic gadgets, display boards, and ICs have been adopted by researchers to improve quality of the end product. Achieving reliable large scale production of nano-systems for skin oriented electronic devices, by addressing issues and limitations is the need of the hour. With a special thrust on recent discoveries in flexible self-healing electronics, which has the capability to transform the scenario of wearable electronic devices, the present review focuses on materials involved, fabrication techniques, challenges encountered and future prospects in the arena of skin oriented flexible electronics.

KEYWORDS: Flexible, wearable, skin like electronics, human machine interface, health monitoring.

1. INTRODUCTION:

Several human skin specific attributes like sensitivity to external stimuli e.g. temperature, strain, humidity etc., providing elasticity to joints and muscles, ensuring safety of delicate body organs by enveloping these and capability of self-healing, act as an inspiration to researchers to mimic these beauties in developing electronic skins/wearable devices for healthcare and other applications [1-3]. Earlier, the installation of rigid functional materials possessing skin-like properties into health monitoring systems rendered these uncomfortable and unfit for long term use thus limiting their utilities. Thus, innovative materials exhibiting such properties which make these compatible with the skin are researched widely. In order to ensure proper functioning of skin-oriented gadgets during both rest and movement situations, a myriad of nanomaterials, which responds to various stimuli, have been developed and incorporated in functional flexible devices. [4]. The main strategy concentrates upon manufacturing techniques to convert relatively heavy and stiff materials into extremely thin, flexible, nano-scale conducting membranes, capable of being printed onto desired substrates [5]. Adopting various design strategies such as serpentine or fractal, dissipation of strain can be accomplished. Although flexible skin-oriented designs have been accomplished using even rigid inorganic nanomaterial [3], however, such gadgets are not free from impediments viz. lesser stretchability and poor mechanical stability. Fig. 1 shows the superiority of flexible materials based sensing devices over the one's developed by involving rigid materials.

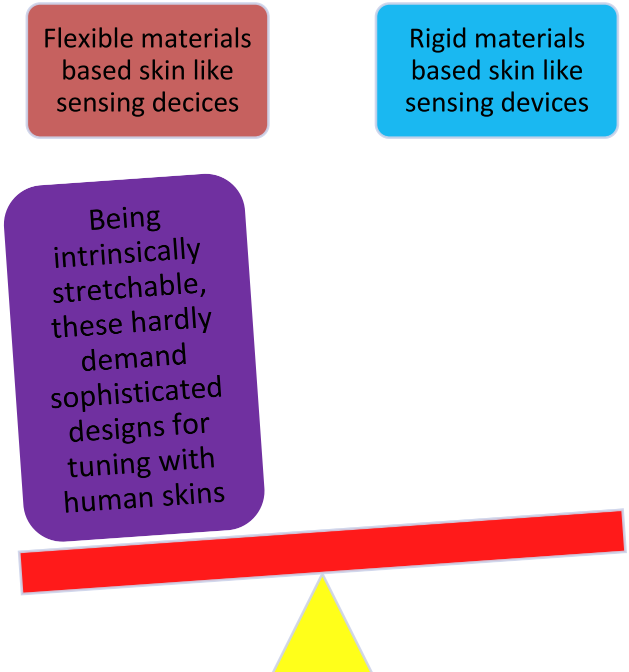

Fig. 1 Flexible materials based sensing devices vs the one's developed by involving rigid materials.

These serious drawbacks have drawn the attention of researchers towards inherently flexible electronics involving active nano-scale materials and/or composites, which are developed by using several novel strategies. Incorporating conductive nano-scale fillers in different polymers to develop inherently flexible and bendable electrodes has been attempted by researchers [6,7]. Being intrinsically stretchable, these hardly demand sophisticated designs for tuning with human skins. Different nano-scale techniques have been developed to carry out efficient strain dissipation into flexible layers, culminating in the development of inherently flexible functional materials for various logic devices, sensor arrays, and display boards [8,9]. Although all these efforts result in the successful development of inherently flexible nano-scale gadgets to copy human skin, these are prone to permanent failure primarily due to fatigue after being repeatedly used, in contrast to human skin which possess the property of self-healing. Researchers across the globe are consequently zeroing on devices which are capable of sustaining functional performance even on being repeatedly used for longer time thus resulting in the fabrication of flexible and rugged skin- oriented electronics [10-13]. Keeping this in mind, in this review, different material combinations and fabrication techniques for developing flexible nanomaterials is under scanner, with a special focus on the recent advances in self- healing electronics [Fig. 2].

2. Conducting Materials Akin to Skin:

Arranging sufficient energy as well as data communication speed for mammoth systems/display units is dependent on the specific resistivity and clarity of flexible conductive materials, that are crucial in small-power multipurpose wearable devices. These requirements must be met under strained conditions also. To cater these requirements, Park and others produced a flexible amalgam of Ag nano scale particles/electro-spun poly (styrene-block-butadiene-block-styrene) (SBS) [14]. The positive ions of silver absorbed by SBS were reduced by hydrazine hydrate, thereby forming

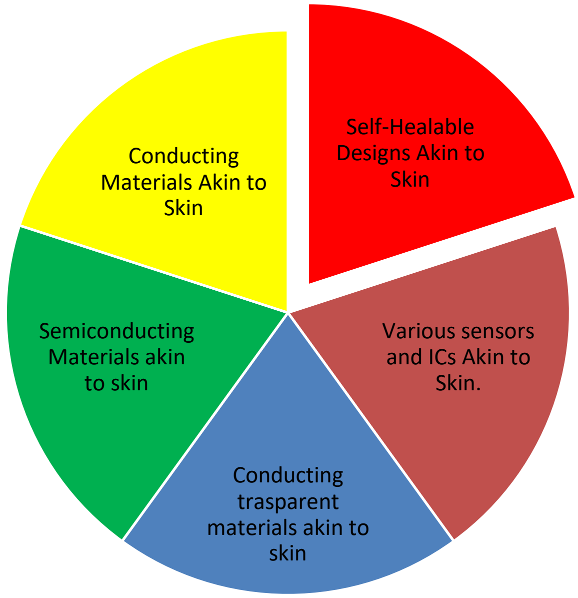

Fig. 2 Different material combinations and fabrication techniques for developing flexible nanomaterials

Ag NPs on an SBS substrate. The flexible amalgam thus formed was stretchable up to hundred percent strain retaining a conductivity value as high as 2.2KS/ cm. In order to obtain enhanced conductivity, Kim and others established self- organized tracks in flexible Au NPs compositions that were developed via two methods viz. layer-on- layer (LOL) formation technique and vacuum-assisted flocculation (VAF) [15]. In PU lined up Au nano scale particles were visible through TEM microscopy. The lined up gold nano scale particles made these films highly conductive (fivefold LOL- 2400 S/cm, fivefold VAF- 35 S/cm) at high value of strains (fivefold LBL-110% strain, fivefold VAF-480% strain). The data implies that non-static nature of the product is likely to impart enhanced flexibility and conductivity to nano- scale ingredients based composites. Matsuhisa et al. fabricated Ag NPs involving micro sized Ag flakes in fluorine rubber [16]. The quantity of Ag NPs was managed by employing surfactant onto the precursor of the amalgam. Surprisingly, Ag NPs were lined up parallel to the strain, enhancing conductivity to the tune of $9.35X10^2$ Scm^{-1} with 0.4K% strain. In spite of all these beauties of these composites, these are not free from limitations as far as wearing for large time period is concerned. Miyamoto and others fabricated flexible and conductive nano-mesh which was permeable to gas [17] by electro-spinning the nano-mesh followed by evaporation of gold film (~70−100 nm) upon it and it was mounted on skin by laminating it upon skin. To dissolve the PVA layer, H_2O was sprayed upon it. No swelling was observed in this process which can be due to the mesh design of the product, which permits flow of sweat through it. This conductive nano-mesh ensured an authentic storage of electromyograms (EMGs) data.

3. Semiconducting Materials Akin to Skin:

Contrary to passive electrical circuit elements, active element such as transistor based circuitry imparts electronic gadgets enhanced spatial resolution, safeguarding specialized operations from unwanted sneak tracks. The crucial components behind this superb functionality are semiconducting materials. Recently, several flexible semiconductors designed for skin-like gadgets have been researched. Kim, Xu and others fabricated ultrathin Si ribbon-like formulations which may be utilized as active switching gadget in logic gates or combined in 3D buckled configurations [3,5,18]. Transfer-printed Si nanoribbon systems designed in a planer geometrical form and inter-faced with serpentine inter-connections possess power of diffusing strains developed locally. Such gadgets prepared by employing elastomeric materials akin to skin, are prepared in the form of laminates and stacked upon the body for performing various measurements. Such devices, however suffer from low density and stretchability. To increase these parameters, inherently stretchable materials have been explored widely. Chortos and others [19] thus developed stretchable totally carbon based transistors consisting of un-sorted CNTs and sorted semiconductors besides utilizing styreneethylene-butadienestyrene (SEBS) as insulator base. These exhibited authentic results with a mean charge mobility of the order of 15.4 $cm^2 V^{-1} s^{-1}$ with an on/off ratio more than 103 and good stretchability. Also, this was noticed that monitoring crucial quantities like limiting surface interface of dielectrics in order to reduce hysteresis as well as bias stress influences are prerequisite for attaining better performing CNTs based transistors. Besides these, inherently stretchable polymeric semiconductors were also fabricated via a specified approach of developing their backbone and side chain. Oh and his team [6] developed a novel architecture for stretchable polymeric semiconductor possessing a capability of diffusing strain at a faster pace. Adopting a new approach, 2,6-pyridine dicarboxamide (PDCA), that possess an affinity of developing H_2 bonding with 3,6-di(thiophen-2-yl)-2,5-dihydropyrrolo[3,4-c] pyrrole-1,4-dione (DPP) polymer backbone, was conceptualized to favor non-covalent non static X-linkage. The developed product exhibited a reliable field- effect mobility of 1.12 $cm^2 V^{-1} s^{-1}$ with hundred percent strain. Xu and his associates noticed nano-confinement assisted by phase isolation between nano-fibril-like semi-conducting (poly (2,5-bis (2-octyldodecyl) -3,6-di- (thiophen-2-yl) diketopyrrolo [3,4-c] pyrrole-1,4-dione-altthieno [3,2-b] thiophen (DPPT-TT) and SEBS elastomer [8]. They exhibited that the nano-confinement successfully decreased the elastic modulus and enhanced ductility thereby empowering the flexible compound to express prominent strain-immune electric parameters i.e. field-effect mobility of the order of 1.32 $cm^2 V^{-1} s^{-1}$ with hundred percent strain.

4. Conducting Transparent Materials Akin to Skin:

In order to make skin-like conductive optoelectronic gadgets a reality, fabrication of transparent stretchable conductors, bears paramount importance [20-23]. Work by Lipomi and his companions [20] indicated that studying morphological variations of stretched mono directional nano-structure mesh will be instrumental for ensuring stretchability as well as clear visibility at the same time. They developed a CNT mesh type flexible structure, having conductivity of the order of 2.2K Scm-1 with one hundred fifty percent strain and possessing transparency more than seventy-nine percent mounted upon an elastomer base. By straining the CNT, a buckled geometry was formed to prepare mechanically robust and electrically conducting electrodes. It was ensured that resistivity remains constant until the afterward strain value remains lesser compared to the strain in the beginning. Liang et al. also embedded Ag nanowires (NW) inside PUA to develop a transparent (>90%), flexible and conducting sheet having resistance of 15 ohms per square with 20% strain, that exhibited good durability during extension/contraction cycles, up to 20% strain [21]. Liu and his team noticed that a special design of "nano-scroll" was created in between graphene sheets during the process of transfer-printing, thereby enhancing stretchability (120% strain) and resistivity of 200 Ω/sq while maintaining high transparency (>88%) [22]. Employing plasticizers has been found to improve stretchability. Besides this, Wang and others mixed ionic liquid (IL) with poly (3,4-ethylene-dioxythiophene): poly (styrene-sulfonate) (PEDOT: PSS) to create superior crystalline nature and improved nano-fiber networks in the product [23]. The developed composites displayed better conductivity of 4.1KScm-1 with 100% strain and 56 Scm-1 with eight hundred percent strain and a transparency level of more than 95%. The presence of IL transforms the PSS matrix into soft material and supply largely acidic negative ions as crucial impurities in PEDOT: PSS, hence enhancing electrical as well as mechanical properties of it.

5. Various Sensors and ICs Akin to Skin:

Flexible sensors have established as indispensable components in skin- electronic circuitry as these, by sensing external stimuli, are capable of transmitting effective data to healthcare personnel on converting it into readable information [2,3,4,24–26]. Strain-tolerant conducting nano compounds are particularly capable of ensuring superb human–machine interfaces [6–7,14–16 ,20-23]. Lipomi and his team synthesized transparent and flexible capacitive sensor arrays, where a low thickness elastomer membrane was sandwiched between upper and lower CNT electrodes [18]. The spread of stress created on application of one mega Pascal pressure to the core part of the mesh was reckoned quantitatively. The researchers advised an optimal balance among clarity and sensitivity, with higher values of strains. Lim and others developed a similar piezo-electric sensor by incorporating CNTs into poly-lactic acid embedded between upper and lower graphene layers [27]. Strain sensitivity of this sensor was enhanced by adding CNT, resulting in a notable enhancement in piezoelectricity of the developed films, on bending. Lee and associates developed a resistive stress sensor comprising of electro-spun conductive nanofibers, involving carbon nano-tubes, graphene, fluorinated elastomer besides an ionic substance in the form of liquid [28]. The nano scale porosity of the product formed ensures superior pressure sensing response and excellent transparency besides the capability to de- couple from strains induced from pressure. The nano-fibrous mesh was combined with a complex system for vast-area pressure-sensing requirements. Manufacturing inherently flexible electronic devices, however requires a complicated procedure. Development of inherently stretchable devices spanning over simple transistors to complex circuits ensures a bright future for skin electronics.

6. Self-Healable Designs Akin to Skin:

Multifunction devices has been developed by incorporating several individual components for the purpose of developing skin electronics for various applications [3,4,24−27]. To mimic real skin, concerted attempts have been made for synthesizing self-healing systems which may restore themselves from fatigue and wear and tear on repeated use [6,11−13,29−31]. Consequently, it is judicious to include here the frontier research in self-healing skin-electronic gadgets which are capable of sensing different stimuli and generating required data ceaselessly even after repeated usage. Working in this direction, Cordier and his team restructured micron sized nickel particles to nano scale [29]. H_2 bonding array in the self-healable product and the nanostructured Nickel particles were instrumental for the mechanical and electrical self-healing capabilities of the product respectively. Under ambient conditions, the self-healing behaviour was attributed to the low glass transition temperature (Tg) of the conducting polymeric amalgam. Based on the composite, Tee and others developed an independent, self-healable product comprising of a supramolecular H_2 bonding array after making some changes in a thermoplastic polymer. They also prepared a self-healing skin capable of sensing a flexion and possessing tactile perception [30]. LEDs assisted by two resistive sensors displayed the variations in flexion angles. These functions are reported to be retained even after a physical damage. Li and his team manufactured a clear, self-healing tactile sensing device having eight by eight electrode network comprising of Ag NW-encapsulated Diels Alder amalgam [31]. Although the developed composite couldn't self-heal on cutting, slight heating up to 353K for half a minute restores its conductivity near to initial value. Huynh and his team designed a multi-purpose self-healing resistor which is able to precisely sense pressure, temperature besides volatile organic compounds (VOCs) [32].

The self-healing capability which takes about a day for completion is attributed to the regeneration of H_2 and covalent di-sulfide bonds in PU. Here, self-healing poly(urea-urethane) thermoset plastic with di-sulfide X-links was employed as a base, and an amalgam of Ag micro-particles and PU plastic was employed as electrode. These electronic components were incorporated on a single platform to make a device which may be utilized in data transfer applications. This compact self-healing architecture may be mounted foe incessant monitoring and treatment purpose.

7. CONCLUSION:

In this review, recent advances in skin-oriented conductors, semi-conductors, ICs, and self- healable devices employed in flexible nano-electronics is described. In spite of the significant progress, the success rate is still limited in comparison to rigid component devices. Selecting nano scale materials and associating these to produce flexible self-healable devices is progressively opening new avenues for potential skin-like electronic nano-systems for applications in biomedical, health monitoring, flexible robotic systems etc. To enhance the feasibility of introducing flexible self- healing materials, in all these sectors, crucial impediments in contemporary skin-oriented electronic systems needs to be identified and resolved. Initially, flexible conductors are required to impart power and transfer data to various electronic parts without injudicious electrical power wastage, even after repetitive use. Precisely, challenges like repeated extension/contraction related fatigue, robustness besides self-healing efficacy of the nano-materials must be addressed. An in-depth study of various issues arising from non- static behaviour of specifically prepared, robust, self-healing, flexible plastics and higher conductance of nano-materials needs to be carried out for identifying and resolving these. Integration techniques to develop long lasting and flexible skin-like electronic nano-systems also needs further improvement. Although innumerous challenges e.g. processing feasibility vis-a-vis developmental methodology, cost of production and yield still stand tall, yet it is expected that involving self-healable ingredients will be an astonishing approach to enable trouble free synthesis of well integrated systems. Transfer-printing methodology is suitable for assembling high-performing individualized self- healing components into super- performance skin-like integrated nano-devices. It, thus can be concluded that a well worked out strategy involving nano-scale non-static nature flexible and self-healing nano-materials will evolve efficient integration techniques to ensure production of robust skin-like electronic nano-gadgets on grand scale with broad applicability.

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