American Journal of Nanosciences
2025, Vol. 9, No. 1, pp. 8-31
https://doi.org/10.11648/j.ajn.20250901.12

Review Article

The Evolution of Self-Healing Electrodes: A Critical Review of Nanomaterial Contributions

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Abstract

The ability of self-healing electrodes to withstand electrical breakdown at high electric fields has drawn a lot of interest to them in recent decades. Applications include electronic skins, sensors, supercapacitors, and lithium-ion batteries have resulted from the integration of conductive nanoparticles in flexible self-healing electrodes. Prior self-healing electrodes based on hydrogels and polymers had low strengths and conductivities. However, nanomaterials offer vast surface area, abundant functional groups, and special qualities that speed up the healing process. Self-healing electrodes, capable of autonomously repairing damage and extending their operational lifespan, represent a paradigm shift in material science and electronic device design. This review paper charts the remarkable evolution of self-healing electrodes, with a particular focus on the pivotal role of nanomaterials in driving this progress. The emergence of self-healing concepts is then discussed, encompassing both intrinsic mechanisms inherent to specific materials and extrinsic approaches that rely on the integration of healing agents. We explore how the distinct physicochemical properties of nanomaterials, such as their high surface area, adjustable conductivity, and catalytic activity, have been used to give electrodes the ability to cure themselves. Specific examples showcasing the successful incorporation of nanomaterials like carbon nanotubes, graphene, MXenes, and metallic nanoparticles into various electrode architectures are presented. The underlying self-healing mechanisms, ranging from reversible chemical bonding to dynamic supramolecular interactions, are elucidated. Furthermore, we critically assess the performance enhancements achieved through nanomaterial integration, including improved mechanical robustness, enhanced electrical conductivity, and extended cycling stability.

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Received: 30 August 2024; **Accepted:** 21 September 2024; **Published:** 23 January 2025

Keywords

Self-Healing, Nanomaterials, Sensor, Electrodes, Energy Storage

1. Introduction

Self-healing electrodes are electrical conductors that, like human skin, have the ability to heal internal damage on their own [1]. By incorporating self-healing material into the electrode, mechanical damage-induced cracks can be repaired, and the device's electrical characteristics can be restored. [2]. Self-healing electrodes have garnered significant interest because of their exceptional capacity to self-heal their damaged areas without the need for additional adhesives, hence regaining their original forms and qualities [3]. According to advancements and research, self-healing active metal alloys or self-healing polymers can be added to electrodes to create self-healing electrodes [4].

Flexible, lightweight, strong, and safe-to-wear electrodes have been developed as a result of the increasing demand for self-healing electrodes [5, 6]. The potential applications of materials with self-healing properties in electronic skins (E-skins), sensors, solar cells, supercapacitors, and other sectors have garnered significant attention [7]. Currently, self-healing electrodes with self-repairing capabilities similar to the skin of human are necessary to improve tenacity [8] Because of these remarkable qualities, the device can continue to work for a long time even if the electrode experiences mechanical cutting, scratching, or cracking. [1]. Numerous self-healing materials have been thoroughly investigated, including ceramics [9, 10], concrete materials, [11, 12] and metals [13]. Unfortunately, the majority of these materials only show a limited number of self-healing cycles and necessitate tedious preparation steps and severe self-healing environments. The connections that are accessible in these materials throughout the healing process include Diels-Alder reactions, Michael additions, metal-ligand interactions (such terpyridine metal complexes), ionic contacts, π-π interactions, and hydrogen bonding [3].

Previous self-healing electrodes composed of hydrogels and polymers have relatively poor conductivities. (<700 S/cm) [14, 15]. Additionally, because of the inherent mechanical characteristics of hydrogels and polymers, most electrodes with self-healing properties have rather low strengths (< 1 MPa) [15]. The electrode may be readily distorted and constrained in everyday applications due to its weak mechanical qualities. Consequently, the need for highly conductive and potent self-healing substances that can restore damaged electrical channels and maintain their electrical properties over time has arisen. Nanomaterials unique functions and huge surface area have led to recent demonstrations of their enormous potential in promoting self-healing [16-22]. The healing process is facilitated by the enormous surface area, numerous functional groups, and unique features of nanomaterials found in electrodes.

However, to the best of the information we have, there aren't numerous reviews that concentrate exclusively on nanomaterial-based self-healing electrodes. For this reason, it's crucial to provide a critical review of nanomaterial-based self-healing electrodes. Therefore, this review provides insight into recent developments in the use of nanomaterials to promote healing and bring novel improvements in self-healing electrodes.

2. Nanomaterials in Self-Healing Electrodes

Due of their potential to create innovative and creative goods across a range of industries, nanomaterials are fasci nating. [23]. Examples of nanomaterials include free-standing nano-objects like carbon nanotubes and nanoparticles, materials having holes the size of nanometers, and other materials with a range of distinct nanoscale characteristics including layered structures that are nanometer thick [24]. The field of nanotechnology is evolving rapidly as it involves the exploration of matter at extraordinarily small scales, typically ranging from 1 to 100 nanometers. The possibilities for nanotechnology are virtually endless, and it could lead to the next wave of scientific and engineering advancement. The materials used in automobiles, buildings, bridges, and aircraft today will give way to stronger, lighter, and longer-lasting nanomaterials in the future. As of now, nanotechnology holds great potential for creating revolutionary instruments to support the creation of a more effective self-healing approach utilizing NPs. Electrodes can profit from NPs' self-healing solutions [25-27].

2.1. Nanomaterials Increase the Efficiency of External Stimuli in Self-Healing

The activation energy required to create covalent bonds or intramolecular interactions is provided by external stimuli like heat, light, and magnetic fields in non-autonomous self-healing. A self-healing process can benefit greatly from external stimuli due to the unique qualities of nanomaterials, such as their ability to convert laser irradiation into heat and

their ability to absorb infrared (IR) radiation. To start the Diels-Alder healing process after infrared radiation, for instance, polyurethane was treated with silver nanowires and graphene nanosheets. [28, 29].

2.2. Nanomaterials Promote Intramolecular Interaction in Self-Healing Electrodes

When nanomaterials are added to self-healing systems, the systems respond to external stimuli more strongly, have better electrical and mechanical qualities, a larger interfacial surface area, and a higher electromagnetic energy conversion efficiency to heat [30]. Functional groups encouraging self-healing through covalent bonds or intermolecular interactions can be attached to nanomaterials' large surface area.

Due to their excellent mechanical properties and surface functional groups such as carboxylate and hydroxyl graphene, carbon nanotubes (CNTs), and graphene derivatives like graphene oxide (GO) have been extensively researched for self-healing applications. Thionyl chloride is often used to convert the carboxylic acid groups on the surfaces of CNTs and GO into more reactive acyl chloride groups, facilitating their functionalization. The desired functional groups are then attached to the surfaces of CNTs and GOs through asymmetrical bonding between acyl chloride groups and furfuryl groups for Diels-Alder reactions, as well as with amine groups for hydrogen bonding. [31]. Furfuryl functionalized multi-walled carbon nanotubes (MWCNT) and furfuryl-modified sty- rene-butadiene rubber (SBR-FS) with bismaleimide under- went a DA reaction to form a composite that was covalently bonded and reversibly cross-linked. Furfuryl functionalized MWCNT in rubber composites showed a dual function that supported and promoted healing. [32, 30].

2.3. Nanomaterial Surfaces and Interfaces Regulates Self-Healing Electrodes

Surfaces and interfaces are commonly acknowledged to dominate and influence the properties of nanomaterials as particle size decreases. However, these surfaces and interfaces' chemical and physical characteristics are typically not quantified or documented. Over the past forty years, surface scientists have discovered many new things regarding surface behavior, such as the significance of both intentional and unintentional surface layers. It takes the same level of diligence to comprehend and regulate the properties of nanoparticles and other nanomaterials as it does to comprehend the chemistry of other surfaces. For example, the electrical properties of Si nanowires are highly dependent on the wire surface and the surrounding environment. [33]. The application and the characteristics of interest can have an impact on the surface nature and impact. Karakoti et al. claim that the significance of surface chemistry of nanoparticles has been greatly underestimated, particularly in relation to toxicological [34].

3. Self-Healing Mechanism

When a self-healing system is damaged, it can use generally accessible resources to restore its functionality or repair itself [35]. Numerous self-healing processes for electrodes have been discovered and published based on the methods. These can be roughly divided into two categories: (i) internal self-healing through reversible linkages, and (ii) exterior self-healing through embedded healing agents (Figure 1).

3.1. Extrinsic (Autonomous) Self-Healing

Extrinsic self-healing, or autonomous healing, is a process in which the mending agent is dispersed throughout the polymer matrix through microcontainers like hollow fibers, micro-capsules, and microvascular networks (Figure 1b). When damage occurs, the shell wall of the microcontainers bursts, releasing the healing agent to seal the tiny and microcracks at the damage location. The distribution pattern, size, shape, and flow of healing agents within micro-containers are a few examples of structural and dynamic factors that influence the materials' capacity for self-healing and for healing. The extrinsic self-healing method's primary flaw is that it can only heal a scratch at one location at a time; further healing is not possible [36]. Vascular self-healing and capsule self-healing are the two forms of extrinsic self-healing.

1. Capsule-based self-healing

The capsule-based self-healing phenomena is the result of the interaction between a catalyst and a microencapsulated healing material within an epoxy matrix. [37]. If there is a break that could occur immediately on the electrode matrix, the microcapsules will instantly rupture, allowing the healing agent to enter the crack by capillary actions By doing this, the healing agent that has been distributed throughout the matrix will polymerize and plug the fracture, stopping it from growing further. The structural composite electrode matrix facilitates this healing process, which does not require external stimulus [38]. The self-recovery mechanism in polymers has also been triggered by the use of micro- or nanocapsules. When the matrix ruptures during self-healing, these capsules repair the injured surface or area [39].

2. Vascular self-healing system

Vascular self-healing occurs when a material is damaged and releases microencapsulated particles of repair resin (hardener/healing agents) into the damaged area. Refillable channels in a polymeric matrix serve as the foundation for the technique. It appears that numerous healing cycles are getting closer thanks to these refillable conduits, which can administer resins in an additional way. This self-healing technology relies heavily on the channels, which serve as both structural reinforcement and repair resin carriers. Up to the point at which the damage initiates self-healing, the channels can be joined in one, two, or three dimensional [40, 41].

3.2. Intrinsic (Non-Autonomous) Self-Healing

The issue of recurrent healing is resolved by the intrinsic self-healing mechanism (Figure 1c). This is a non-autonomous method that involves introducing reversible crosslinks into the polymer matrix. These crosslinks cause the healing mechanism to activate in response to environmental cues, including heat, light, pH, and moisture. The same wounded area might recover multiple times due to the reversible chemical linkages. Reversible in temperature Diels Alder (DA) reactions are widely employed in the synthesis of several thermosetting polymers with self-healing properties. The two functional groups that function as diene and dienophile in Diels Alder reactions most frequently are furan and maleimide. Because of the DA reaction, DA crosslinks are formed at lower temperatures. At high temperatures (retro-DA temperatures), these weak DA crosslinks can break, resulting in smaller chain fragments that reflow more readily to the site of injury. As a result, the crack may close and the polymer may be repaired within a certain temperature range as the chain segments may be reunited by DA processes [42]. Chemical contacts primarily consist of covalent bonds, while physical interactions arise from the development of van der Waals forces [43, 44].

3.3. Role of Nanomaterials in Self-Healing Mechanism

When carbon nanotubes are added into nanomaterials, it may result in reversible hydrogen bonding interactions in electrodes [45]. As a result, the self-healing phenomena is dependent on damage repair via bond formation. Aside from physical interactions, functional carbon nanotubes may form covalent connections with electrodes [46]. Furthermore, depending on their particular roles, carbon nanotubes can act as an electron giver or acceptor in relation to electrodes for the production of covalent bonds. [46].

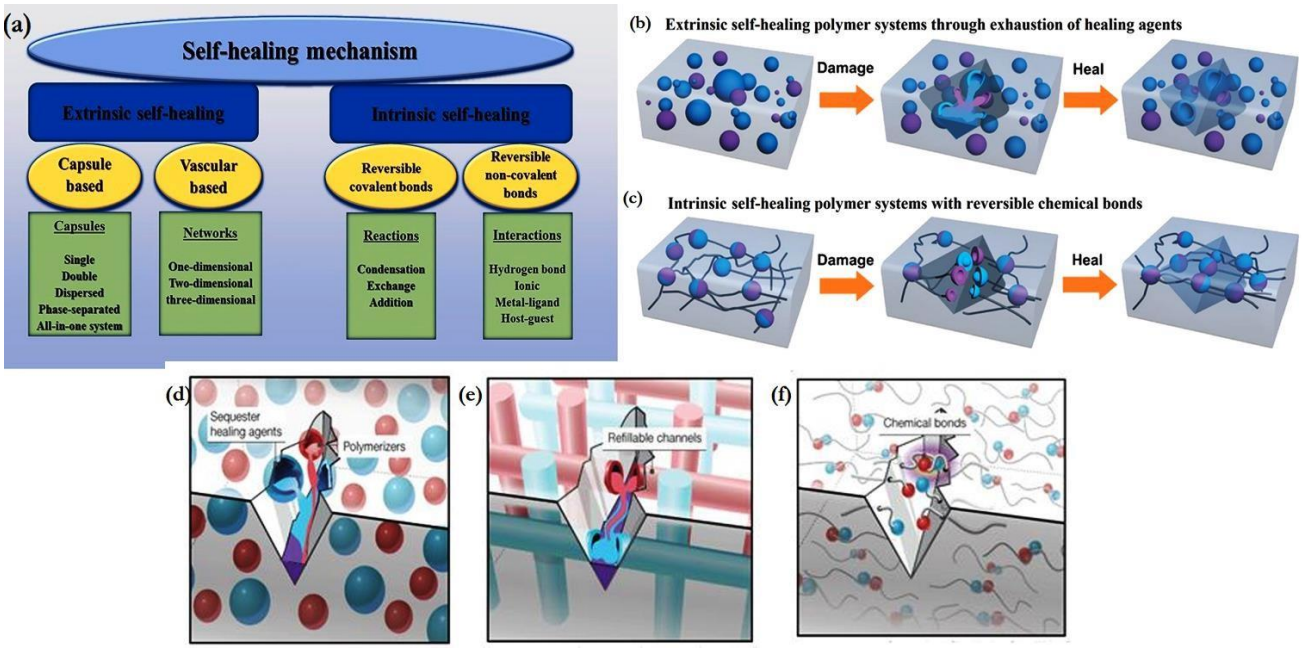

Figure 1. *(a) Graphical representation of self-healing mechanisms adapted from [4] (b) Extrinsic self-healing mechanism (c) Intrinsic self-healing mechanism adapted from [47] (d) Self-healing agent based on capsules (e) Vascular self-healing agent (f) Intrinsic self-healing agent retrieved from [37].*

4. Characterization Techniques

To fully comprehend the self-healing process and its applications, nanomaterial characterization is a crucial first step. This section focuses on the many ways for characterizing nanomaterials used in self-healing electrodes. Because of the nanoscale size of the sample, deep structural characterization of nanomaterials is difficult; in reality, nanoparticles are made up of only a few atoms, and more conventional characterization techniques may be ineffective. As a result, new techniques for nanomaterial characterization are needed to comprehend the relationship between changes in atomic ordering, modifications, and (degradation of) performance [48]. The use of characterization techniques on electrodes postmortem or after cycling is essential to enhancing battery performance.

They are able to pinpoint the specific failure mechanism at play and offer remedies. Because electrode combinations (binder, active material, and electron conductor) are often so complex, even an accurate single spectrum could be inter-

preted incorrectly. For instance, it can suggest a species interaction even when certain species are too far away to interact, or it might result in the composition of a compound even though two compounds should be taken into consideration [49]. The detailed descriptions of the following analytical instruments are used to characterize nanomaterials:

1) Field emission scanning electron microscopy (FESEM) analysis

This method targets an electron beam at the sample holder using an electron cannon. A vacuum is created in the chamber to direct the electron beam toward the incident material while the electron gun is mounted atop the apparatus. Lenses and an electromagnetic field in the device regulate the electron beam's path. A sample is struck by an accelerated electron beam, which causes some of the electrons to be backscattered, some to pass through it, and some to emit secondary electrons. The detector tracks these scattered and secondary electrons. Finally, a computer can be used to record the sample's morphology [24].

2) Auger electron spectroscopy

Auger electron spectroscopy (AES) collects data on specific nanomaterials by using an incoming electron beam with spatial resolution. Because AES uses electron excitation, concerns regarding electron transmission through a particle and backscattering could lead to some excess sample parts being excited to produce Auger electrons, hence reducing the true spatial resolution. [50]. When collecting AES data from a feature visible in a scanning electron image that can be accessed in an AES apparatus, analysts often presume that the entire signal is created from the area where the electron beam strikes. When an electron beam passes through a nanoparticle or other nanosized structure below or into the surrounding material, auger signals are produced and recorded from both the nanofeature and the surrounding area. AES has shown to be particularly useful when paired with other techniques to offer information on nanostructured materials when the potential challenges are acknowledged [24].

3) Scanning probe microscopy

Scanning probe microscopy (SPM) is widely recognized for its ability to characterize nanostructures, in contrast to conventional surface analysis tools. AFM and scanning tunneling microscopy (STM) are two popular examples of SPM-based techniques. AFM is able to capture a three-dimensional image of nanoparticles dispersed on a flat surface. Additionally, it can provide quantitative and qualita tive data on the size, shape, surface roughness, and other physical characteristics of nanoparticles in certain circumstances. AFM-generated images can be used to measure par ticle-size distributions and even particle volumes, although it is important to use caution when addressing possible tip artifacts. Even though most high-value nanoparticle testing uses isolated particles on flat surfaces, attempts are being made to broaden the applicability range. [24].

Numerous research groups have been able to creatively increase the applicability of AFM devices because to their relative affordability. Using nanoparticles (or molecules) linked to scanning probe tips, one can evaluate the interactions of individual nanomaterials (or molecules) with a flat surface or with other nanoparticles [51]. These measurements enable the investigation of interaction forces in a range of environments and offer information on them. Using AFM, the surface roughness of nanoparticles has also been measured. This is really beneficial because most surface roughness measurement techniques are inapplicable to small, curved surfaces such as these. Furthermore, the surface roughness of ceria nanomaterials has been examined using AFM and TEM to examine the impact of particle size and production methods. [52, 53].

4) Fourier transform infrared spectroscopy (FTIR)

The technique known as Fourier Transform Infrared (FTIR) monitors the absorption of electromagnetic radiation at wavelengths between 400 and 4000 cm^{-1}. It is a method for getting an infrared spectrum of a solid, liquid, or gas being absorbed or emitted. A molecule becomes IR active when it absorbs infrared light, which alters its dipole moment. The placement of bands linked to the kind and strength of bonds as well as certain functional groups can be seen in a recorded spectrum, which can be used to learn about interactions and molecular structures [24].

5) Brunauer-Emmett-Teller (BET) analysis

Since nitrogen interacts substantially with most materials and is often available in high purity, it is frequently used in BET surface area studies. Due to the relatively limited interaction between gaseous and solid phases, detectable levels of adsorption are achieved by chilling the surface with liquid nitrogen [24].

6) Ultraviolet-visible spectroscopy analysis

One low-cost characterization approach that is frequently used in nanomaterials research is UV/Vis spectroscopy. It contrasts the light reflection intensity from a reference material with the light reflection intensity from a sample. The formation of NPs is ascertained by measuring the optical absorbance spectra. The transmission and absorption characteristics of the sample can be determined using UV-Vis spectroscopy. NPs' optical properties are sensitive to concentration, shape, size, aggregation state, and refractive index close to the NP surface. These materials can be identified, investigated, and characterized using UV-Vis spectroscopy. It can also be used to assess the stability of NP colloidal solutions [23].

7) X-ray diffraction (XRD) analysis

A common method for describing nanomaterials is X-ray diffraction (XRD). Chemical information for elemental and phase analysis is also provided. When assessing a variety of materials, such as fluids, powders, and crystals, X-ray diffraction (XRD) is an essential technique [54].

8) Transmission electron microscopy (TEM) analysis

For nanomaterial analysis, the TEM instrument is frequently utilized. TEM can provide information on bulk ma- terials at magnifications ranging from extremely low to high since it operates on the concept of electron transmission. The various morphologies of nanomaterials are examined using this technique. Particle size, shape, and grain size are all accurately

measured by TEM. The filament in a TEM is the instrument's source of electrons; it might emit electrons in the form of a thermionic or field emission filament. To attain high energy, the filament's produced electron is accelerated by an electrode or cathode [24].

9) Raman Spectroscopy

Raman spectroscopy investigates chemical structure, phase, polymorphism, crystallinity, and molecular interactions using vibrational modes. In this method, light from a laser source accidentally hits sample molecules, interacting with them through inelastic scattering and chemical reactions to reveal the identities of the molecules. A bond's specific vibration mode, such as C-C, C-H, C=C, N-O, and so on, is represented by each peak in a Raman spectrum [55].

5. Applications

In recent times, self-healing electrodes have garnered significant interest due to their ability to self-heal after injury or illness. Considerable effort has been put into developing new self-healing materials that satisfy the needs of many applications. Remedy chemicals can be stored in a variety of materials, including concrete, ceramics, metals, and polymers. This method calls for complex procedures for the medicinal ingredient's dispersion and encapsulation. More significantly, the therapeutic content of the capsules will quickly run out, limiting how frequently it may be administered. Another helpful method for producing materials with self-healing qualities is reversible dynamic bonding. [56].

Self-healing electrode nanocomposites have demonstrated intriguing qualities as novel, long-lasting, safer, and sustainable materials for energy storage devices, [57-61] flexible electronics [62, 63], actuators [64], structural components and coatings that prevent corrosion [65]. Many intriguing applications, such as electric-skin modifications, have been made possible by the integration of conductive nanoparticles onto flexible self-healing electrodes [66, 67]. Electrodes' self-healing nature allows them to withstand electrical breakdown even at relatively high electric fields. Furthermore, because these self-healing electrodes are compatible with standard photolithography and wet etching, high-resolution patterning for device fabrication is made possible [68].

5.1. As Energy Storage Devices

Devices that store and convert energy electrochemically, such as batteries, fuel cells, supercapacitors, H_2O/CO_2 electrolysis, and so forth, are essential to the global low- and zero-carbon energy strategy for sustainable development. They also help meet the increasing demands of a wide range of applications, from behind-the-meter uses to mobility [69, 70]. Nonetheless, certain devices' capabilities, operational stability, life cycle, stability, and efficiency are still insufficient.

Materials used in self-healing electrochemical energy devices are capable of identifying and fixing some or all external or internal damage on their own. One characteristic of next-generation smart energy storage systems is their ability to cure themselves. due to its ability to work intelligently even in the face of mechanical, thermal, or radiation damage while maintaining its original integrity [71]. The creation of electrode materials with self-healing properties has been the main goal of efforts to date. For example, certain self-healing materials have advanced from using microencapsulated substances or micro-hollow fibers to intrinsic molecular healing with recurring reversible healing [72].

Dynamic bonds that are reversible, like hydrogen bonds, dynamic metal-ligand interactions, hydro-phobic interactions, dynamic covalent boroxine bonds, and so on, have been the subject of recent research [63, 73, 74, 75]. Despite advancements, only a small number of self-healing material types have been successfully used in ESDs. This is because using self-healing materials in ESDs requires additional requirements, including processability, electrochemical and thermal stability, mechanical, electrical, and self-healing properties [76].

5.1.1. Lithium-Ion Batteries (LIBs)

Lithium-ion batteries (LIBs) have been used to power a wide range of devices, including electric cars, implanted medical equipment, portable electronics, and stationary power plants. Further development for high-performance has focused on the exploration of innovative materials including silicon and its composites, as well as metallic alloys, in order to achieve high energy capacity and extended cycling life. However, throughout the charge and discharge processes, electrochemical reactions in batteries lead to structural alterations from mechanical microcracks, which reduces cycling life [77-79]. Lithium-ion batteries (LIBs) are leading the secondary battery industry due to qualities including minimal self-discharge, high voltage, extended cycle life, and high energy density (150-250Wh kg-1). They are commonly found in a wide range of portable electronic devices, including cell phones, digital cameras, laptop computers, and electric cars. [76].

Electrode materials experience mechanical fracture as a result of the battery's repeated ion insertion and extraction. By mimicking the self-healing properties of biological systems, electrode composites can significantly increase the lifespan of energy storage devices and restore the related electrochemical performance [80]. With a primary focus on the healing of their material structural and mechanical properties, a large number of studies on lithium-ion batteries (LIBs) have been carried out based on the advantages of self-healing electrodes to increase their dependability, cycle life, and lower production costs. To address this, special self-healing electrodes for lithium-ion batteries (LIBs) have been created that use liquid metal's reversible solid-liquid transformation to seal cracks during the lithiation and delithiation cycling process. To create a robust solid-electrolyte interface (SEI) layer, the synthesis of self-healing polymeric binders using dynamic bonding to restore conductive networks during volume change is also being studied [81]. Polymer electrolytes have

developed to the point that they may self-heal after being sliced, folded, or stretched without affecting the battery's electrochemical function [82].

The first self-healing flexible LIB was presented by Zhao et al. [58]. After being aligned on a self-healing substrate, CNT sheets were loaded with $LiMn_2O_4$ and $LiTi2(PO_4)_3$ to form the electrodes. The gel electrolyte and separator were composed of aqueous sodium carboxymethylcellulose and lithium sulfate. The structure and electrochemical characteristics of the as-fabricated LIB may self-cure in a matter of seconds. The capacitance dropped after the sixth cutting, going from 28.2 to 17.2 mAh g^{-1} at 0.5 A g^{-1}. Furthermore, strong mechanical self-healing was shown by the LIB. Liquid metals and their alloys have garnered significant attention due to their exceptional electrochemical characteristics, high electronic conductivity, and deformability, particularly in the energy storage industry.

In order to achieve self-healing through the reversible solid-liquid transition of low-melting-point liquid metals during lithiation and delithiation cycling processes, gallium (Ga) was employed as a liquid metal negative electrode. The researchers found that during lithiation, the Ga electrode crystallized and changed into a solid electrode. The solid-state Ga electrode underwent delithiation and was subsequently transformed back into liquid, which remarkably repaired the electrode's cracks. Cycling led to a significant reduction in capacity, which was brought on by the top of the electrode producing SEI during lithiation. The produced SEI layer consumed Li^+, electrode material, and electrolytes during delithiation and was unable to be dissolved [77].

The fragmentation and fracture leading to lithiation-induced volume change in the electrode, especially in the first cycle, has hindered the use of silicon (Si) for high-capacity anodes of lithium-um-ion batteries [83, 84].

One approach to enhancing capacity loss and cycle performance in Si anodes was to thoroughly analyze the morphological and structural variations in Si in order to comprehend the self-healing mechanism of Si-Al anodes.

Bhattacharya *et al.* [83] used in-situ optical microscopy and micro-Raman spectroscopy to study self-healing Si-Al electrodes. In conclusion, they discovered that the Si-Al interface cracks closed at the end of the cycling process and suggested that a composite electrode made of Si particles buried in a ductile phase could improve the batteries' electrochemical performance.

A liquid metal-based self-healing LIB anode was demonstrated by Wu et al. [85]. This liquid metal was stabilized by an RGO/CNT skeleton and consisted of a Ga-Sn liquid metal alloy. Ga-Sn alloy is self-healing and can be stored as a liquid at 25 ℃ since Sn reduces the melting temperature of Ga. During repeated cycling, the RGO/CNT skeleton prevented Ga-Sn aggregation or separation and enhanced electrode conductivity. During charge and discharge, the Ga-Sn grew increasingly abrasive. Ga-Sn displayed a solid-state with bumps and ravines at full lithiation, which is explained by volume expansion. The flat surface morphology was recovered after delithiation. This conduct avoided both expansion and contraction. Consequently, during 4000 cycles, the LIBs based on liquid metals demonstrated good cycle stability. This work suggests a novel approach for long cycle life LIBs.

5.1.2. Supercapacitor (SCs)

Another significant class of electrochemical ESDs with properties like LIBs in energy storage are supercapaci- tors (SCs). Their extended cycle life of over 100,000 cycles and high specific power of over 10,000 W kg-1 make them ideal energy sources for electric vehicles, lasers, and rockets. [86]. Redox or electrical double layers provide the basis of the energy storage mechanism of solar cells. SCs are well known for their quick charge and discharge times, high power density, and prolonged cycling performance. In real-world applica- tions, the ability to self-heal can significantly increase their stability and service life. The current focus of research on self-healing SCs has primarily centered on mechanical and electrical repair up to now. Of self-healing SCs, the two main elements influencing their production are their electrodes and gel electrolytes. [2]. The first self-healing SC with mechanical and electrical features was proposed by Wang et al. [87].

Restoring ionic conductivity is the goal of a self-healing dielectric. Polyelectrolytes are perfect materials for self-healing electrolytes because of their excellent compatibility with electrode materials, strong ionic conductivity, and adjustable mechanical characteristics. Huang *et al*. reported (Figure 2e) a dual crosslinked polyelectrolyte for self-healing SC [88]. CNT paper coated with polypyrrole (PPy) was em- ployed as an electrode. The self-healing polyelectrolyte was made using vinyl hybrid silica nanoparticles (VSNPs-PAA) and double-crosslinked hydrogen bonding in polyacrylic acid. The synthetic SC made with this polyelectrolyte maintained 100% of its capacitance even after 20 healing cycles. Lin et al. created a novel ferric enhanced dual physical crosslinking polyelectrolyte (Fe-DPCL) by employing acrylic acid as the hydrophilic monomer and stearoyl methacrylate as the hy- drophobic monomer. [89]. Excellent ionic conductivity (>30 mS cm-1) was exhibited by the H_2SO_4 in Fe-DPCL, and the hydrogel as a whole showed good stretchability and self-healing capabilities. Even after the seventh injury, the built SC (Figure 2f) demonstrated good self-healing capabili- ties and capacitance retention of approximately 86%.

In another study, [61] generated a highly robust, multiresponsive healing polypyrrole incorporated gold nanoparticle/CNT/poly(acrylamide) (PAM) (GCP@PPy) hydrogel electrode with a hierarchical honeycombed network structure by growing PPy nanoparticles in the GCP framework. The electrode exhibited remarkable stretchability (elongation of 2380%) and new Au-SR bond-triggered optical and electrical healing (94% healing efficiency) after being chemically crosslinked with gold nanoparticles. By sandwiching multi- functional GCP@PPy electrodes with a CNT-free GCP (GP) hydrogel electrolyte and chemically

soldering a layer of Ag nanowire (AgNW) film to the hydrogel electrode as the cur- rent collector using a combination of spray coating and NIR laser irradiation, a stretchable and real-time omni healable all-gel-state super-capacitor device was assembled as a proof-of-concept demonstration. This stretchable superca- pacitor exhibited the highest energy density (123 Wh cm-^{2}) and areal capacitance (885 mF cm-2) of any known superca- pacitor. The M (M = Au, Ag)-SR bond-induced integrated configuration and the intrinsic stretchability and heal ability of the electrode and electrolyte produced a high capacitive sta- bility with 89.5% of the initial value restored at a superhigh device-level strain of 800%, as well as rapid optical healing performance over ten healing cycles. A maximum of 92% of the specific capacitance was recovered throughout the charge-discharge process, marking the first example of an electrical real-time omni-healable supercapacitor prototype. These advantages confirm that this supercapacitor is the best performing multifunctional supercapacitor among those that have been reported, offering a wide range of potential applications in the flexible electronic devices

5.1.3. Capacitors

Recently, [90] has published a study on the dynamic rebuilding of a conductive nanostructure Following the film's separation from the substrate, the conductive CNT network was primarily discovered on the film's upper surface. They discovered that after the aforementioned embedding procedure, the resistance of the CNT network increased tenfold, most likely as a result of some insulating polymer interpenetration between the CNTs. They also proposed that an increase in electrical resistance could be the outcome of doping the electrode with Au salt. The author first investigated the quantitative harm produced by a razor blade using in situ monitoring in order to assess the self-healing properties of a carbon nanotube electrode. Subsequently, they examined the effects on the CNT electrode material at different time intervals of different forces (0.54 N) and repetitive damage (20 times of a 2 N force). As demonstrated by the results, the CNT electrode at the injured area was able to mend itself (Figure 2 a, b, c). Significantly, the self-healing polymer matrix demonstrated remarkable promise for network restoration, paving the way for longer-lasting electronic applications down the road.

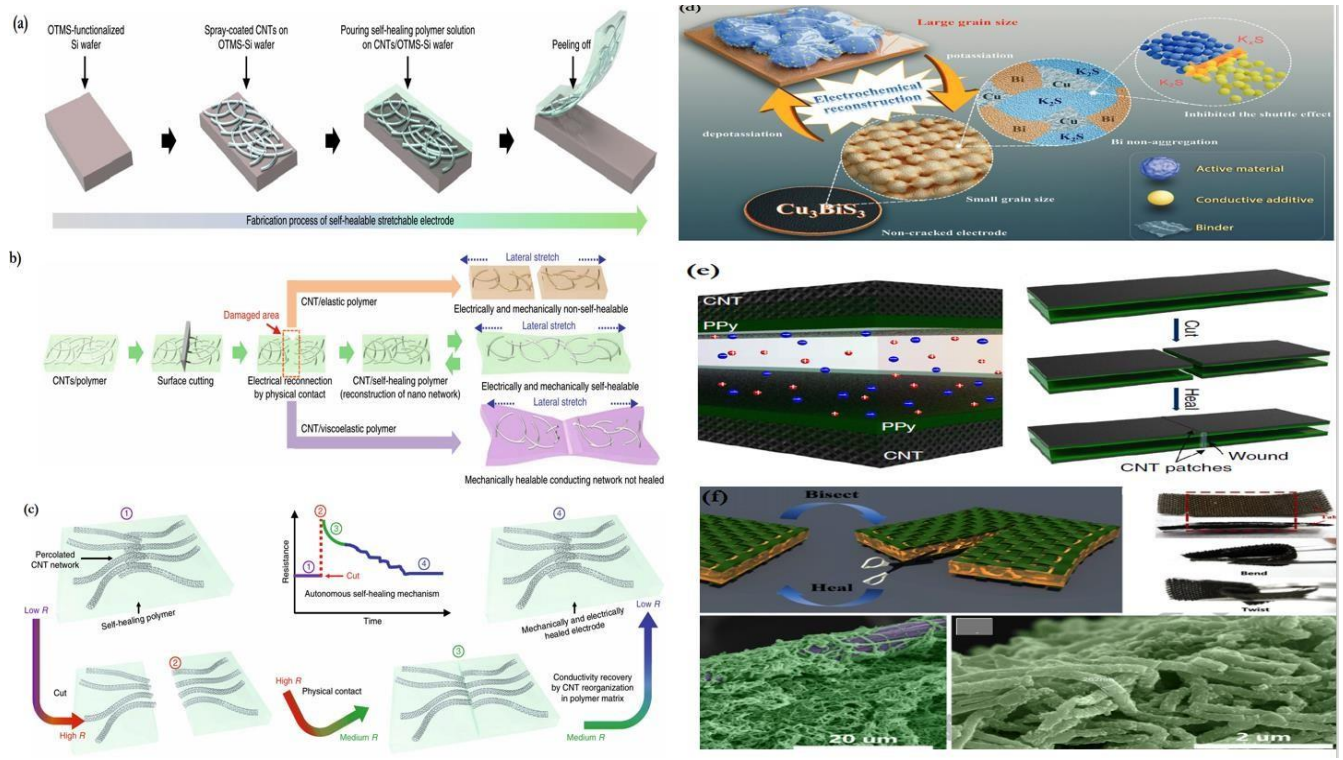

Figure 2. *Recent self-healing electrodes used as energy storage devices (a) The fabrication of a self-healable stretchable electrode by embedding a carbon nanotube (CNT) conductive network into a self-healing polymer matrix is depicted schematically (b) Sequential schematic illustration of several substrate types and their responses to damage and reconstruction (c) Carbon nanotubes incorporated in a self-healing polymer matrix are proposed for recuperation. The reconstruction of the CNT network is based on the following observations: (1) continuous resistance reduction over time, (2) stretchability recovery, (3) keeping good conductivity with strain for recovered electrode, and (4) lack of evident physical separation in the CNT network in the recovered cut region. Because of the dynamic nature of the self-healing polymer, the electrode was electrically healed autonomously, allowing the CNT network to rearrange and restore both stretchability and conductivity Recovered from [90]. (d) The self-healing process and the effect of the Cu3BiS3 nanocrystal potassium ion electrode are depicted schematically retrieved from [80]. (e) Dual crosslinked polyelectrolyte based self-healable supercapacitor (f) Ferric enhancing dual physical crosslinking polyelectrolyte based self-healing supercapacitor recovered from [2].*

Anode material with self-healing properties for potassium-ion hybrid capacitors (PIHC) was presented by [80] in a different investigation. Cu_3BiS_3 anodes can exhibit ultra stable cycling performance, preserve their original properties after potassium and depotassium, and have both phase and morphological reversibility, in contrast to binder designs. The reversible electrochemical rebuilding during continuous charge/discharge operations is advantageous for maintaining the material's structure and function. Additionally, there were two advantages to the conversion events that took place throughout the charge and discharge process: Cu can prevent Bi nanoparticles (NPs) from clumping together, improving the electrochemical performance and long-cycle stability of the Cu3BiS3 electrode. As a result, not only does the Cu_3 BiS_3 electrode have a long cycle life in half cells, but it also has a cycle life of 12000 cycles in PIHC complete cells. The self-healing process is as follows (Figure 2d): when discharged to 0.01 V, the big grained Cu_3BiS_3 NPs change into small-grained intermediates (Cu, K_3Bi, and K_2S).

When charged to 1.6 V, the Cu/Bi heterostructure contact is formed, which can lessen the shuttle effect. Moreover, Cu can be used as a separator to stop significant Bi particle agglomeration, which enhances Cu3BiS3 kinetics. Ultimately, the small-grained intermediates revert to large-grained Cu_3BiS_3 NPs when charged to 3V. The self-healing method allows for the reversible repair of Cu3BiS3 NPs and is dependable in repeating itself over a period of cycles, up to 2000 cycles, thereby, greatly relieving electrode material cracking caused by ion intercalation. It can attain superior quality and a longer cycle life because to its self-healing feature. The Cu_3BiS_3 electrode has a 2000-cycle cycle life and a steady capacity at 500 mA g^{-1}.

5.2. As Sensors

In the past, sensors were made of a variety of materials, including semiconductors, conducting polymers, and metals. [91, 92] But the stretchability of the majority of conventional strain sensors is quite low. Furthermore, because it is less sensitive, hence it is not wholly acceptable for usage with the human body [93]. Stretchability strain sensors can be of several types, depending on aspects such as sensor type, polymer nature, conducting nature of nanomaterials utilized, and fabrication method. Polymer composite-based strain sensors typically have two components: polymer and conductive electrode [94]. The conductive electrode generates a resistance or capacitance output signal when physical strain changes, while the polymer offers flexibility and stretching properties. Moreover, the polymer component guards against damage to the conducting channel of the sensor electrode during strain testing. Covering a strain sensor that is affixed to the skin can generally help to prevent direct contact with human fluids and environmental factors that may hinder or interfere with the ability of the sensor to detect resistive or capacitive signals. [95].

Interestingly, a number of investigations have been carried out to examine different nanomaterials, including graphene, carbon black (CB), carbon nanotube (CNT), gold nanowires (AuNWs), silver nanowires (AgNWs), and silver nanoparticles (AgNPs), for the purpose of fabricating strain sensors in order to satisfy the essential need for sensors. [96, 97]. Comparable flexible polymers, such as silicone elastomer, rubber, latex, and polydimethylsiloxane (PDMS), have been employed as the substrate in recent investigations. [98]. Table 1 illustrates different self-healing electrodes recently used as sensors.

Furthermore, in order to overcome the rigidity and susceptibility of conventional metallic electrodes, [99] developed an ionic hydrogel with high deformation tolerance and quick self-healing ability. Additionally, the researchers demonstrated a skin-mounted, stretchable, self-healing device known as a Triple S active sensor (TSAS), which operates on the principles of electrostatic coupling and electrostatic induction. With a maximum output voltage of 78.44 V, a detection limit of 0.2mN, a quick response time of 1.03 m/s, an excellent signal-to-noise ratio, and outstanding long-term service stability, the skin modulus-matched TSAS has remarkable sensing capabilities. During arm muscle training, joint dexterity of bending angle and functional signals from the triceps brachii and biceps muscles can be collected simultaneously with TSAS. Another option is to wirelessly send the signal to a terminal for analysis. TSAS's high sensitivity, dependability, affordability, and ease of use make it a promising next-generation method for assessing muscle function in real-time during rehabilitation training.

Furthermore, [90] employed electrodes with self-healing properties that were produced as active parts of apparatuses. Specifically, a three-electrode wearable self-healing ECG sensor was made (Figure 3a, v). The sensor was affixed to the human arm securely (Figure 3b). The self-healing electrode's damage recovery was demonstrated by the recovery of real-time cardiac signal recordings (Figure 3a, vi). The authors found that, in the event of injury, the signal may quickly return to its typical baseline. They also indicated that EMG measurements may be made using these instruments. Furthermore, employing PDMS-MPU0.4-IU0.6 dielectric (thickness 500 m), they created a capacitive strain sensor that was positioned between the top and bottom self-healing electrodes. The strain sensor (Figure 3a, vii) demonstrated consistent strain sensing capabilities even after sustaining damage (up to 25% strain) due to the material's homogeneity and the strong H-bonding connection that forms between the self-healing electrode and dielectric. Additionally, a notch was added to the strain sensor in order to inspect any fracture propagation brought on by straining (Figure 3a, ix). It was surprising that it could be stretched to 50% strain without the cracks spreading any farther.

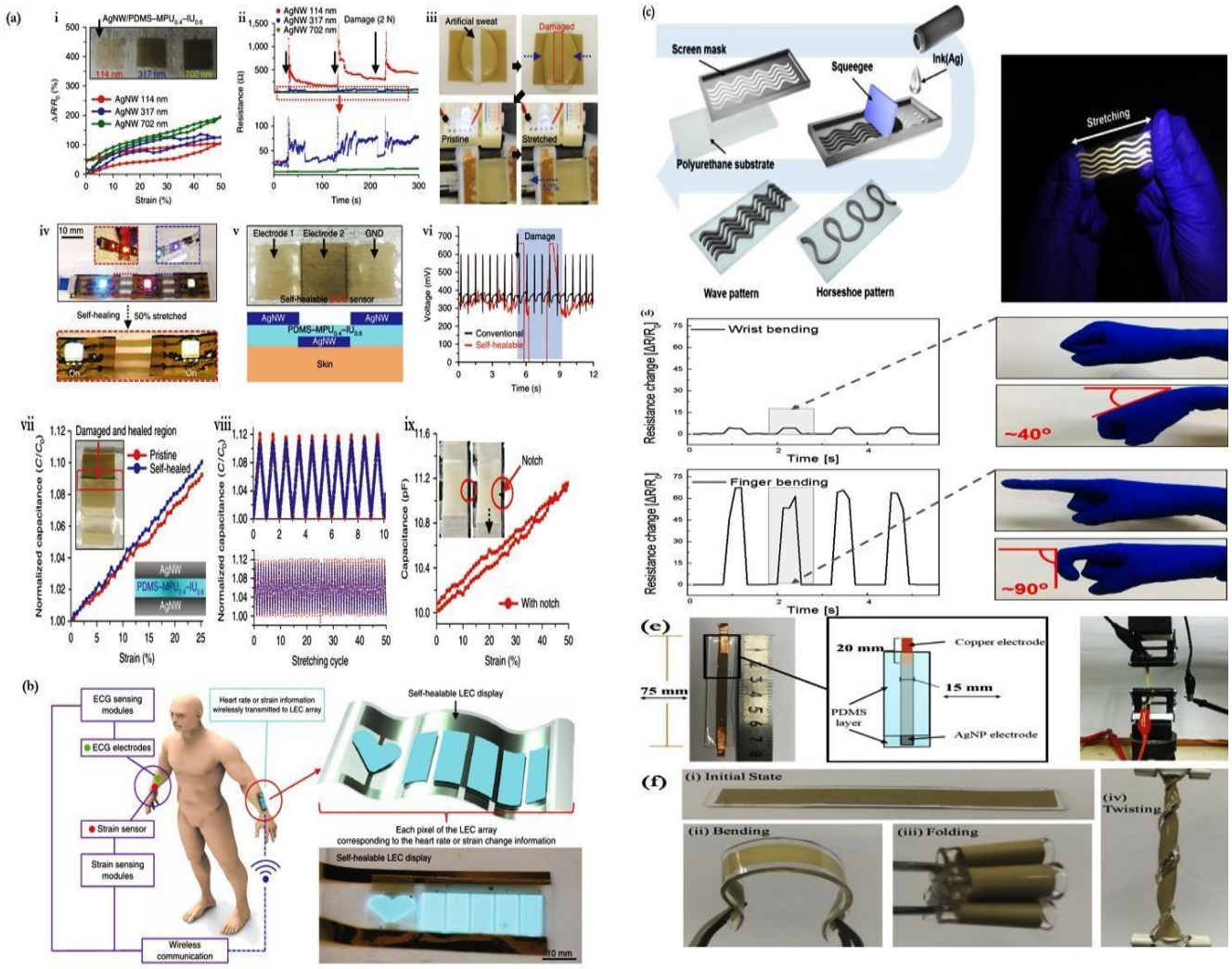

Figure 3. *Recent self-healing electrodes used as sensors (a) i, Plot of relative resistance change of AgNW with three different thicknesses as a function of strain ii, The top frame displays a plot of resistance of a damage-resistant electrode with varying AgNW thickness as a function of time while making three successive cuts (2 N each), while the bottom frame shows a magnified low-resistance range. iii, Images (top two frames) of the sequential self-recovery process of a self-healing AgNW/PDMS-MPU0.4-IU0.6 composite electrode even in the presence of an artificial sweat solution. Following the autonomous healing process, the healed electrode is stretched to 50% strain iv. Photographs (top and left, partial cut; top and right, entire cut; bottom, self-healed) demonstrate self-healable interconnects interfaced with commercial LEDs and passive modules on polyimide substrates. Autonomously healed 1st interconnect is stretched well to deliver electrical power to 2nd LED cell (bottom). v, Self-healing ECG sensor image and associated schematic. Cardiac signals from the self-healing ECG sensor (red) and a commercial ECG sensor (black) are exhibited for comparison. The blue highlighted portion demonstrates how the electrode pads were damaged and then recovered in a matter of seconds. vii, Comparison of strain sensitivity and viii, stretching cyclic (30% strain) test before (red) and after (blue) making a partial incision on the strain sensor. The damaged strain sensor is shown in the inset. ix, Because of the high toughness of the polymer substrate, which is capable of preventing fracture propagation, the strain sensor with a notch (insets) still demonstrates good stretchability without any electrical loss (b): Image showing attachment of the fabricated ECG sensor attached to the human arm Adapted from [90] (c): Schematic screen-printing process for patterning of wavy and horseshoe-shaped Ag NP electrodes on stretchable polyurethane substrate and Stretched wavy patterned Ag NP electrodes on polyurethane substrate (d) Image showing the Ag NP-based strain sensor on a human wrist and human fingers which was bent about 40^0 and about 90^0 which is about 20% and 40%strain respectively Adapted from [100]. (e) optical photograph, dimension, tensile measurement of strain sensors (f) picture of strain sensors in (i) initial, (ii) bending, (iii) folding, and (iv) twisting states Adapted from [105].*

In a different study, [100] used simple screen printing to create flexible, reasonably priced Ag nanoparticle (NP) self-healing electrodes on polyurethane (PU) surfaces. To create the stretchable electrodes, they simply designed the Ag NP electrode in a horseshoe-shaped and wavy pattern as a function of line width. At room temperature, the stretchable electrodes had low sheet resistances of 1.64-2.85 Ohm/square. As seen in Figure 3c, there was a continuous resistance change up to 15% (horseshoe pattern) and 20% (wavy pattern) strain on the screen-printed Ag NP electrodes with a 3-mm line width.

Comparing the 15-20% stretchability to other stretchable fabrics, it is fairly low. Stretchability for inorganic Ag NP electrodes is 15% to 20%, which is acceptable and rather high for the production of electronics that can be stretched Furthermore, with critical inner and outer bending radii of less than 1 mm, the screen-printed Ag NP electrodes may be employed as extremely flexible electrodes. The high flexibility and stretchability of screen-printed Ag NP electrodes on PU substrate was explained by a tenable stretching mechanism. Additionally, they employed stretchable Ag NP electrodes for strain sensors and stretchable interconnects for light-emitting diodes. Ag NP electrodes show great promise as a stretchy electrode for wearable and flexible sensors given that they can detect human motion (Figure 3d).

Recent research has shown that cellulose nanofiber/MXene and flexible, self-healing ionic hydrogel electrodes can be used for bionic and sensing applications [101, 102]. Bionic devices rely on flexible conductive materials for the transmission and collection of biosignals. One of the most important biological signals is body temperature, so it's imperative to create a wearable body temperature sensor that can provide accurate temperature readings in real time [103]. Conductive materials that sense temperature by thermoresistive behavior caused by thermally-induced charge carrier scattering or charge transport enhancement are among the most promising body temperature sensing methods [104]. The resistance of the thermoresistive temperature sensor material should not fluctuate owing to mechanical stress since resistance changes produced by various curved surfaces and human body movements make acquiring correct signals challenging.

Again, employing conductive silver nanoparticles (AgNPs) on a polydimethylsiloxane (PDMS) substrate, [105] produced resistive-type strain sensors. Using a drop-casting approach, AgNPs covered PDMS strain sensors (Ag-PDMS) and AgNPs modified PDMS composite strain sensors (Ag@PDMS) were created. These strain gauges had a sand- wich like appearance. The electromechanical performances of Ag-PDMS and Ag@PDMS composite strain sensors were assessed, and the effects of AgNPs loading with PDMS at 0.20 wt%, 0.25 wt%, and 0.30 wt% were examined. With a gauge factor of 10.08 over a 70% strain range, a strain sensor made with 0.25 weight percent AgNPs loading in PDMS (Ag0.25@PDMS) composite shown good stretchability, du- rability, and repeatability in addition to a fast time response. This study's Ag0.25@PDMS composite strain sensor is ca- pable of detecting finger motion, suggesting applications for highly sensitive strain sensing, such as human body motion capture.

Furthermore, [106] developed, tested, and produced an elastic, self-healing, damage-sensing skin structure. The skin was built layer by layer using polyimide sheets, an ultraviolet (UV)-curable epoxy, and substrate made of copper-clad polyimide. UV-curable epoxy was applied as a structural adhesive and as a self-healing fill material. An assortment of LC circuits, every possessing an individual resonance frequency, was connected to the skin configuration. When skin is injured, UV-curable epoxy is secreted and is cured by sunlight. Furthermore, damage affects one or more of the LC circuits, changing the resonance frequency.

An integrated antenna coil was used to find and identify the damaged skin. A 2 2 array of LC circuits in a proof-of-concept skin is shown. The test findings demonstrate the effectiveness of fault isolation and the skin's ability to mend itself.

5.3. Electronic Skin

Electronic skin is a term for an electronic system that can replicate the stretchability, self-healing capacity, and sensory range of human skin. It can also convert a variety of environmental inputs, including pressure, humidity, and deformation, into electrical impulses [107, 108]. Among the numerous applications for e-skin are robotic prosthetics, wearable electronics, artificial intelligence, and soft robotic systems [109-111]. With the aid of self-healing properties, e-skin's stability and longevity may be improved.

Cao *et al*. utilized an amorphous and chemically compatible polymer to construct a stretchy and self-healing e-skin for aquatic conditions (Figure 4a). [112] The e-skin's ionic conductivity can be adjusted to 103 S cm^{-1}. The device continues to function normally even when it reaches a peak of 2000%. Because e-skin's ion-dipole interactions are so reversible, it can heal itself in both wet and dry environments.

After ten cuts, the electrical conductivity's healing efficiency was 90.7%. The skin demonstrated the sensory capabilities of its potential uses by being incredibly responsive to touch, pressure, and strain.

Pan et al. revealed that a bionic tactile plastic e-skin based on a composite hydrogel exhibited excellent sensitivity for strain detection (GF = 14.14), remarkable super stretchability (>5000%), and a rapid rate of self-healing (3 s, 95.73%). The hydro-gel-based e-skin (Figure 4b) is capable of detecting human ECG and electromyography signals; it has potential uses in prosthetics and real-time healthcare [113].

In a study, [1] combined silver nanowires (AgNWs) with liquid crystal graphene oxide (LCGO) to create a printable, durable, and electrically conductive electrode. It was simple to make the conductive ink by mixing the AgNWs and LCGO solutions. Several freestanding shapes, like as a butterfly, can be produced using the minuscule (3m-thick) electrode. The LCGO/AgNW composite exhibited the maximum conductivity and strength of 17,800 S/cm and 4.2 MPa, respectively. These values were 24 and 4 times greater than those of a self-healing electrode that had been manufactured before. When exposed to ambient temperatures and moisture, the LCGO/AgNW composite self-healed, regaining 95% mechanical toughness and 96.8% electrical conductivity over the undamaged state. The composite's electrical characteristics exhibited metallic tendencies. Furthermore, the researchers demonstrated that the LCGO/AgNW electrode may be used in light-emitting diodes (LEDs) and other electronic circuits

(Figure 4c). These findings imply that the composite could be utilized to sense compression and temperature as an artificial electronic skin (Figure 4d). Potential uses for this self-healing artificial electronic skin include human condition monitoring and robotic sensing system electrodes.

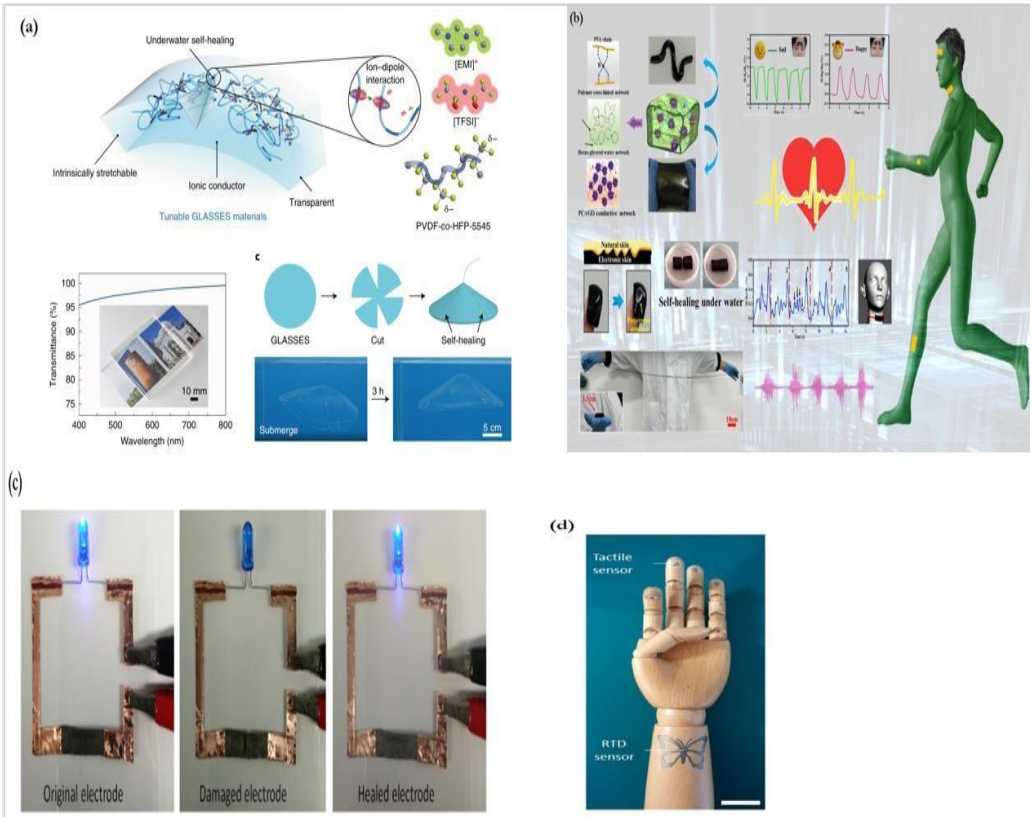

Figure 4. *Recent self-healing electrodes used in electronic skin (a) A gel-like, aquatic, stretchable, and self-healing e-skin retrieved from [112] (b)An hydrogel-based self-healing e-skin Retrieved from [113] (c) Photographs of the electrode within a circuit with an LED before and after breaking and after healing (d) An optical image of the artificial e-skin showing e tactile sensor and resistance temperature detector were located on the fingers and wrist, respectively retrieved from [1].*

5.4. Other Applications

Recently, a semi-transparent poly-tetrafluoroethylene (PTFE)/AgPdCu (APC)-PTFE-based superlattice electrode was produced [114] by sputtering ultra-thin APC-PTFE hybrid layers over a polyurethane (PU) substrate in alternating directions at room temperature (Figure 5a). Modifying the PTFE thickness and the number of layers in the stacked superlattice allowed for the optimization of the superlattice electrodes' mechanical and electrical characteristics.

A sheet resistance of 8.5 Ohm/square and high conductivity even at 100% strain are displayed by the top PTFE/APC-PTFE/PTFE/APC-PTFE/PTFE superlattice electrode (Figure 5b). The exceptional ability of the superlattice electrode to stretch and maintain its conductivity stems from the robust, three-dimensional network formed by the conductive APC material within the flexible PTFE polymer matrix. Moreover, putting the superlattice electrode under external strain changed the optical transmittance, which might be exploited in stretchy electronics for optical switching. The superlattice electrodes can be used to create wearable temperature sensors, stretchable thin-film heaters, and stretchable electroluminescent devices (Figure 5c). The successful demonstration of these stretchy devices, enabled by the remarkable stretchability of the superlattice electrode, highlights the immense promise of this manufacturing technique for the development of next-generation wearable and flexible electronics.

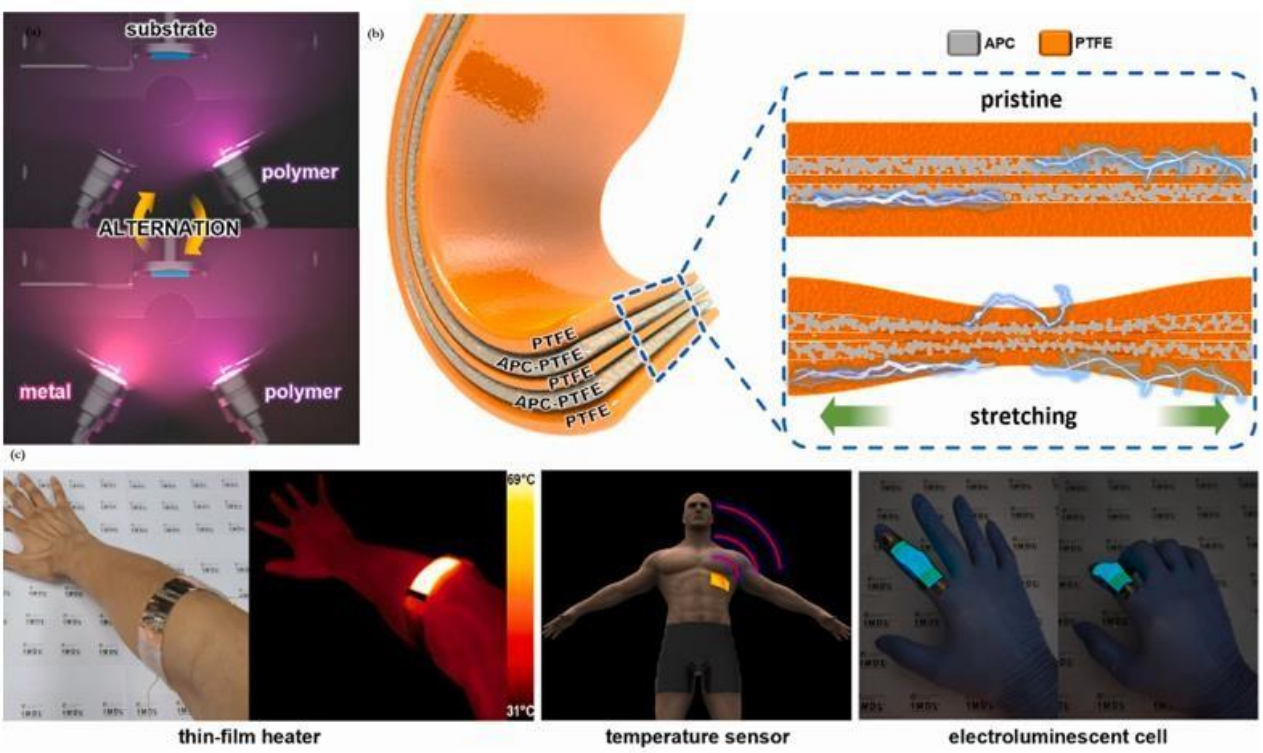

Figure 5. *Structure and potential uses of superlattice electrodes. (a) Illustration of the superlattice electrode fabrication procedure employing a multi-cathode sputtering device. (b) Superlattice electrode structure with PTFE/APC-PTFE/PTFE/APC-PTFE/PTFE. (c) Promising su- perlattice electrode applications include a stretchable heater, stretchable temperature sensors, and flexible electroluminescent cells Adapted from [114].*

Table 1. *Self-healing electrodes applied as Sensors.*

Material	Substrate	Fabrication Method	Resistance	Stretchability	Application	Ref.	Future use
AgNPs	PU	Screen printing	1.64-2.85 Ohm/square	15-20%	Strain sensor	[100]	wearable & stretchable sensors
AgPdCu (APC)	PU, PTFE	Magnetron sputtering	8.5 Ohm/square	100%	stretchable heater, temperature sensors, and electrolumines- cent cells	[114]	Stretchable & wearable elec- tronics

Material	Substrate	Fabrication Method	Resistance	Stretchability	Application	Ref.	Future use
AgNPs	PDMS	Drop-casting method	-	70%	Strain sensor	[105]	human body motion capture
AgNPs	PDMS	Single-step direct transfer process	6.67 ×10-6 Ω·m	25%	Strain sensor	[115]	flexible pressure sensor, wearable human motion detection devices
AgNPs	PDMS	A pattern-template-induced printing	-	0-5%	Strain sensor	[116]	monitoring of delicate defor- mation for human facial expression
GnP	PU	Layer by layer lami- nating method	1430S/cm	25%	Strain sensor	[98]	Wearable devices

Material	Substrate	Fabrication Method	Resistance	Stretchability	Application	Ref.	Future use
Graphene	PDMS	SLG film	0.42S/cm	20%	Strain sensor	[117]	-
MWCNTs	TPU	Fused deposition modeling	0.01S/cm	100%	Strain sensor	[118]	-
Copper	polyimide sheets	Layer-by-layer technique	-	-	Skin sensor	[106]	-
Elastomer, MWCNTs	PDMS, PBS	Dynamic dual network	2.9×10^{-4} S/cm	20%	Rate sensor	[119]	Smart sensing materials field
AgNWs	LCGO		17,800 S/cm				

TPU= Thermoplastic polyurethane; MWCNTs=multiwalled carbon nanotube; PDMS = poly (dimethylsiloxane); PBS= polyborosiloxane; LCGO=Liquid crystal graphene oxide

6. Challenges and Limitations

6.1. Characterization Challenges

A lot of nanomaterials are challenging to identify, which bothers scientists and engineers who want to employ them for energy, biomedical, or other uses, as well as regulatory agencies that have to know how they affect the environment and public health [120]. The lack of comprehensive nano-material characterization has far-reaching consequences, hindering the reproducibility of research, creating roadblocks in scaling up production for industrial applications, and making it difficult to accurately assess potential risks to human health and the environment. Although most research groups aspire to conduct high-quality research, comprehensive nanomaterial characterization is not achieved for a variety of reasons. First, a number of sectors and researchers involved in nano-materials research are unfamiliar with the special preparation and analysis issues that nanoparticles present. Furthermore, the complexity of nanomaterial characterization often demands a multidisciplinary approach, posing a challenge for research teams who may lack access to the full spectrum of specialized techniques needed to acquire a comprehensive understanding of these materials. Adding to these challenges, the diverse data required for a thorough understanding of nanomaterials often necessitates specialized instruments and analysis methods that may be unfamiliar to research teams. This knowledge gap can lead to suboptimal execution of critical characterization steps or misinterpreta- tion of the data, ultimately hindering progress. Researchers are only now starting to recognize some of the limitations of analytical approaches that have been successfully used in other disciplines. Nanoscience and nanotechnology are still in their infancy [121].

6.2. Challenges of Stability of Nanomaterials

Nanomaterials' high interface-to-volume ratio is an inherent property. As a result, the energy required to form an interface is high, reducing the stability of nanostructures in comparison to similar large-scale materials. This is especially important in harsh conditions such as intense laser radiation, high temperatures, and oxidative environments [122]. These materials' and systems' capacity for self-healing helps them prevent this issue by reducing structural and functional issues. However, it is impractical to apply recognized processes for micro- or macro-sized materials because of the severe size limits. Therapeutic medications can be encapsulated and transported via channels in both macroscopic and nanoparticle forms. Nonetheless, the small size of nanoparticles limits encapsulation possibilities [123]. Indeed, the stability of conductive nanomaterials based on met-al is still a source of concern. Moreover, sintering is required for the surface deposition of metallic nanoparticles. In order to create a contin- uous thin sheet, the liquid phase carrier solution must be re- moved from the substrate during the sintering process [124].

6.3. Challenges of Cost and Scalability

The majority of intrinsically flexible nanocomposite electrodes made from nanomaterials such as metal NW and elastomer matrix require complex manufacturing processes like pre-straining, transfer, patterning, or solution processing. Many of these methods are not cost-effective for commercialization since they are not appropriate for today's vacuum-based display and semiconductor industries. Furthermore, there is minimal performance enhancement when stretchable electrodes are used as device components due to the high contact resistance between heterogeneous fillers like metal NW and other components. It is therefore necessary to create a revolutionary production process or a conduction filler to replace the metal NW. For creating stretchable electrodes, vacuum-based techniques like sputtering and evaporation are the most effective coating procedures due to their high degree of consistency and dependability [125, 126].

Similar fissures and pulverization of the self-healing elec-

trode are produced by the threefold volumetric expansion of silicon during lithiation and contraction during delithiation. This causes a loss of electrical contact and excessive growth of SEI, which ultimately results in capacity degradation [127]. Even when metal alloys or polymer binders are added to silicon, cycling still causes cracks to form. Research on how Si diameters affect capacity delay has shown that capacity decay may be enhanced by particles with a diameter of 150 nm and nanowires with a diameter of 250 nm [128]. However, the preparation process's difficulties and cost of scalability limit its industrial applicability. Self-healing polymers covered with silicon microparticles could produce better volumetric energy density while also being less expensive and easier to scale-up. Despite these efforts, it is necessary to use strong adhesive polymer binders that enable for simple ion passage in order to sustain extreme mechanical stress during numerous cycling improvements [129].

6.4. Environmental Concern

The poor cycling performance of a Si anode electrode is shown in Figure 6 as a result of the electrode's extreme shrinkage and volume expansion during cycling, which fractures the electrode and leads to electrical isolation of the Si particles and electrode delamination from the current collector. Significant structural degradation of the S cathode promotes uncontrollable diffusion of the intermediate polysulfides, which leads to low Coulombic efficiency (CE), a reduction in cycle life, and rapid capacity fading. Batteries fail catastrophically due to direct electrode contact caused by the simple fragmentation of polymer electrolytes during complex deformation during charge and discharge. The self-healing characteristics seem to be essential since they can enhance the battery's cycle stability by naturally mending cracks and fractures [130]. However, because the materials used are susceptible to environmental factors like oxygen and water, there aren't many known successful cases [131].

To mitigate lithium dendrite formation, the electrolyte interface (SEI) layer plays a crucial role in preventing unwanted reactions between the electrolyte and lithium. However, continuous cycling leads to frequent volume changes and detrimental interactions between the electrodes and electrolytes, resulting in the development of an unstable and thick SEI layer. This layer consumes more lithium ions and electrolytes within the battery, lowering its Coulombic efficiency (CE) and shortening its cyclic life. Artificial SEI layers, which cover the surface of the Li metal to function as a physical barrier and stop the lithium ions from coming into direct contact with the electrolyte, have been made using liquid metal alloys. However, because these manufactured SEI layers lack the mechanical ability to withstand constant volume fluctuation during continuous cycling, damage and separation result [132-134].

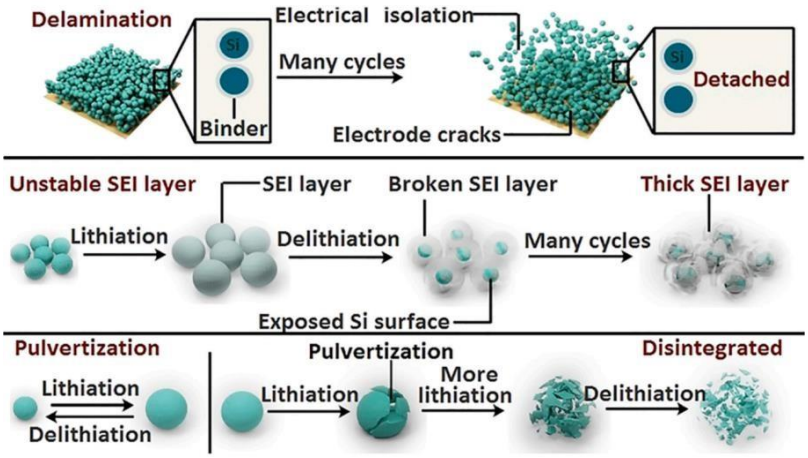

Figure 6. *Schematic Illustration of a few electrode malfunction occurring over the course of several cycles [135].*

7. Future Prospects

Significant progress has been made in this important field of research thanks to extensive investigation on self-healing electrodes. Self-healing qualities have been achieved by incorporating nano-scopic processes into various materials. Conversely, there are still few reports of self-healing nanostructures. But over the past ten years, intensive research has led to a much better understanding of the underlying ideas and mechanisms in addition to the development of a number of novel self-healing electrodes (even up to the creation of new commercial goods). This knowledge will eventually be used as the basis for the creation of novel methods and distinctive self-healing electrodes. These materials will mostly be microscopic and macroscopic, but the process of self-healing may also rely on nanoscopic occurrences and distinct compounds. Nonetheless, new self-healing nanomaterials will continue to be developed, with consideration

given to their special qualities. Future research will focus more on the significance and applications of nanomaterials, which will increase the need for these materials to self-heal because of their special properties that make them brittle. Remarkably, nanomaterials' ability to self-repair seems to be one step ahead of "normal" materials' ability to heal in terms of restoring particular traits other than mechanical qualities.

The current focus of macroscopic materials' self-healing is primarily on mechanical property restoration. There have only been a handful published exceptions thus far. However, other qualities, including optical capabilities, are often restored or preserved as a result of nanomaterials' ability to cure themselves. Research on nanoparticles can thus open the door to a greater understanding of macroscopic properties.

More research on stretchable transparent electrodes is necessary for the commercialization and practical application of self-healing electrodes, despite the fact that recent advancements in the development of self-healing electrodes using nanomaterials have led to the demonstration of several stretchable electronics application examples. The first and most important step is to develop workable processes for mass-producing high-quality nanomaterials while guaranteeing their chemical and thermal stability. To address a specific material's weaknesses, hybridization studies with other nanomaterials are also necessary. The most important, affordable, and wide-spread production processes for integrating these electrodes with substrates or other functional layers ought to be researched [136].

Self-healing materials are still in the early stages of development, but these cutting-edge materials could be useful for solving problems in a range of applications. For instance. It is projected that in the future, industries will be the only uses for these materials. Because of this, there are currently very few successful industrial applications, mostly in the automotive, aerospace, and structural industries [137].

8. Comparative Analysis

The nanomaterials utilized in the self-healing electrode can be broadly categorized into three groups: (i) conductive polymers [140], (ii) carbon-based nanomaterials [139], and (iii) metallic nanomaterials [138]. In addition, a novel family of materials known as transition metal carbides and nitrides (MXene) has been used more frequently in self-healing electrodes recently because of its improved environmental stability, dispersibility, and conductivity [141]. One of the most intriguing possibilities for self-healing electrodes is a nanomaterial made up of a flexible elastomer, conductive polymers, graphene, nanowire (NW), metal nanoparticles, and carbon nanotubes [142-144].

Metal NW, a deformable conductive filler, performs the best among the other conductive fillers. The many kinds of nanomaterials utilized in self-healing electrodes are contrasted and compared in this section. Additionally, it evaluates their efficacy and fit for a range of applications.

Table 2. *Showing comparison of nanomaterials used in self-healing electrodes in terms of their nature and areas of application.*

Nanomaterial	Nature	Application	Ref.
Copper and Copper oxide	Toxic	-	[145]
Carbon Nanotubes	toxic	Smart robotics	[146]
Graphene/ Graphene oxide	toxic	-	[147, 148]
Silver nanoparticle	toxic	-	
Nanoclays	safe	organo-electronics	[149]
Mxenes	toxic	Biomedical applications	[150]

Low-dimensional bulk metal nanostructures in the form of zero-dimensional (0D) nanoparticles (NPs) or one-dimensional (1D) nanowires (NWs) are known as conductive metal nanomaterials. Copper (Cu), gold (Au), and silver (Ag) are common metal-based functions. The most interesting substitute among these are silver-based nano-materials (Ag NPs and Ag NWs) [151]. Ag NPs and Ag NWs have always been preferred over Au NPs due to their higher cost and necessity for high temperature and high vacuum during deposition, even though gold nanoparticles (Au NPs) are resistant to oxidation and corrosion and have conductivity comparable to silver nanomaterials. Researchers are drawn to silver-based nanomaterials because they offer the highest conductivity of any metal, a low rate of oxidation, and the advantage of simple displacement in the liquid phase [152, 153]. Due to their similar conductivity to silver nanomaterials, affordability, and durability in the presence of oxygen and moisture, copper nanoparticles (Cu NPs) have shown great promise as a better functional material in recent years [154]. However, the formation of a sur-

face oxide layer during at- mospheric synthesis poses a difficulty because of the ther- modynamic instability of Cu NPs [155]. Metallic nanoparti- cles, such as AuNWs and AgNPs, have obviously piqued the interest of researchers working on flexible sensors [156, 157]. AgNPs, which have high electrical and thermal conductivity, improve the performance of stretchy sensors. Furthermore, AgNPs are less expensive than other metallic nanoparticles such as gold nanoparticles (AuNPs) and iridium nanoparticles (IrNPs) [158]. There have recently been reports of con- structing an electrode with a stretchability of 250-460% uti- lizing extremely long AgNW or CuNW wires [159, 160]. It is also reported that AgNW coated woven mesh can be stretched up to 20-40% [161]. According to the findings, metal NW has a low percolation threshold because to its large aspect ratio and superior ductility, making it beneficial for obtaining high stretchability and high conductivity simultaneously. Lithi- um-ion electrodes, superconductors, catalysts, and other commercial products can all be made with copper and copper oxide nanoparticles. It is known that as Cu-based nanoparti- cles age in different environments, their characteristics will change over time, despite the fact that numerous studies have suggested that they might be hazardous. Research on the effects of Cu nanoparticles in various chemical states has been carried out [145, 162]. This includes assessments of the bio- logical reaction to Cu-based nanoparticles that are supplied with a known surface chemistry. One-dimensional carbon nanotubes (CNTs), two-dimensional graphene, and zero-dimensional carbon quantum dots (CQDs) are examples of carbon-based nanomaterials that span all dimensions. Carbonaceous nanoparticles are widely used because of their excellent mechanical qualities, increased electrical and thermal conductivity, and environmental durability [163]. Graphene is a two-dimensional planar structure made up of a single layer of carbon atoms. It possesses exceptional elec- trical and mechanical qualities, making it one of the most promising materials in flexible electronics [164]. Since GO is not electrically conductive and cannot be used to fabricate conductive electrodes, its dispersibility comes at the expense of conductivity. Known as single-walled (SWCNT) or multi-walled (MWCNT) carbon nanotubes, carbon nanotubes are the cylindrical or tubular form of a single or multi-layer concentric carbon atom or graphene sheet [165]. A CNT's chirality along its graphene sheets determines its properties, which can be metallic or semi-conducting. While CNTs' conductivity is comparable to that of metal, their resistance is increased by synthesis-related defects [166].

9. Conclusion

In this review, we focused on the advancements made recently in self-healing electrodes based on nanomaterials and their recent applications. Similar to human skin, self-healing electrodes have the capacity to mend internal damage and return electrical conductivity. Because of their enhanced electrical and mechanical characteristics, enhanced interfacial surface area, enhanced responsiveness to external stimuli, and enhanced electromagnetic energy conversion efficiency, nanomaterials are essential to self-healing systems. Nano- materials have progressed to the point that flexible, lightweight, robust, and safe electrodes may now be created. These electrodes have use in solar cells, supercapacitors, sensors, electronic skins, and other fields. Due to their distinct properties and high surface area, nanomaterials have shown great promise in fostering self-healing, allowing devices to function optimally over extended periods of time even in the face of damage. However, there are still few reports on self-healing nanomaterials. Under severe circumstances, the stability of nanostructures is restricted by inadequate characterization and elevated interface-to-volume ratios. The high interface-to-volume ratio of nanomaterials, particularly under adverse conditions, diminishes the stability of nanostructures. For self-healing electrodes to be commercialized and put to practical use, more research on stretchable transparent electrodes is needed. Given the unique properties of these materials, the creation of novel self-healing nanomaterials will continue. Future uses and the importance of nanomaterials will make self-healing qualities more important for these materials, whose unique characteristics render them susceptible to damage.

Abbreviations

CNTs	Carbon Nanotubes
GO	Graphene Oxide
DA reaction	Diels-Alder
SBR-FS	Styrene-Butadiene Rubber
MWCNT	Multiwalled Carbon Nanotube
MWCNT/SBR	Multiwalled Carbon Nanotube / Styrene-Butadiene Rubber
FESEM	Field Emission Scanning Electron Microscopy
AES	Auger Electron Spectroscopy
SPM	Scanning Probe Microscopy
STM	Scanning Tunneling Microscopy
AFM	Atomic Force Microscopy
TEM	Transmission Electron Microscopy
FTIR	Fourier Transform Infrared Spectroscopy
BET	Brunauer-Emmett-Teller
UV	Ultraviolet-visible
NP	Nano Particles
XRD	X-ray Diffraction
H_2O/CO_2	Water/Carbon(iv)oxide
LIBs	Lithium-ion Batteries
Ga	Gallium
Si-Al	Silicon-Aluminum
Si +	Silicon Ion
Li +	Lithium-ion
SEI	Solid Electrolyte Interface
SCs	Supercapacitors
VSNPs	vinyl Hybrid Silica Nanoparticles

Fe-DPCL	Ferric Improved Dual Physical Crosslinking Polyelectrolyte
VIacid	H_2SO_4 - Tetraoxosulfate
PIHC	Potassium-Ion Hybrid Capacitors
AgNW	Silver Nanowire
Cu_3BiS_3	Copper Bismuth Sulfide
Cu	Copper
1D	One-dimensional
PDMS	Polydimethylsiloxane
CB	Carbon Black
NPs	Nanoparticles
TSAS	Triple S Active Sensor
EMG	Electromyography
LEDs	Light-Emitting Diodes
LCGO	liquid Crystal Graphene Oxide
PU	Polyurethane
0D	Zero-dimensional
PTFE	Polytetrafluoroethylene
NW	Nano Wire
IrNPs	Iridium Nanoparticles
CE	Coulombic Efficiency
GF	Guage Factor
Au NPs	Gold Nanoparticles

Acknowledgments

The authors also wish to thank researchers who have previously contributed to this field of study.

They apologize if any prior work has been unintentionally overlooked or inadequately cited.

Conflicts of Interest

The authors have no known personal conflicts of interest.

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