

Review

Multifunctional Polymeric Nanocomposites for Sensing Applications—Design, Features, and Technical Advancements

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Abstract: Among nanocomposite materials, multifunctional polymer nanocomposites have prompted important innovations in the field of sensing technology. Polymer-based nanocomposites have been successfully utilized to design high-tech sensors. Thus, conductive, thermoplastic, or elastomeric, as well as natural polymers have been applied. Carbon nanoparticles as well as inorganic nanoparticles, such as metal nanoparticles or metal oxides, have reinforced polymer matrices for sensor fabrication. The sensing features and performances rely on the interactions between the nanocomposites and analytes like gases, ions, chemicals, biological species, and others. The multifunctional nanocomposite-derived sensors possess superior durability, electrical conductivity, sensitivity, selectivity, and responsiveness, compared with neat polymers and other nanomaterials. Due to the importance of polymeric nanocomposite for sensors, this novel overview has been expanded, focusing on nanocomposites based on conductive/non-conductive polymers filled with the nanocarbon/inorganic nanofillers. To the best of our knowledge, this article is innovative in its framework and the literature covered regarding the design, features, physical properties, and the sensing potential of multifunctional nanomaterials. Explicitly, the nanocomposites have been assessed for their strain-sensing, gas-sensing, bio-sensing, and chemical-sensing applications. Here, analyte recognition by nanocomposite sensors have been found to rely on factors such as nanocomposite design, polymer type, nanofiller type, nanofiller content, matrix–nanofiller interactions, interface effects, and processing method used. In addition, the interactions between a nanocomposite and analyte molecules are defined by high sensitivity, selectivity, and response time, as well as the sensing mechanism of the sensors. All these factors have led to the high-tech sensing applications of advanced nanocomposite-based sensors. In the future, comprehensive attempts regarding the innovative design, sensing mechanism, and the performance of progressive multifunctional nanocomposites may lead to better the strain-sensing, gas/ion-sensing, and chemical-sensing of analyte species for technical purposes.

Keywords: nanocomposite; polymer; interaction; strain-sensing; gas-sensing; chemical-sensing



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1. Introduction

Nanocomposites have been applied as efficient materials for sensing applications [1]. The chemical structure of nanomaterials greatly influences sensing properties [2,3]. In the case of polymer nanocomposites, a range of polymers from the categories of conducting polymers, thermoplastics, elastomers, etc., have been used [4–6]. Among all polymers,

conducting or conjugated polymers have been found effective to be utilized to design competent nanocomposite sensors [7,8]. Conjugated polymers like polyaniline, polypyrrole, polythiophene, and conducting blends have been mostly used for the sensing applications. The inclusion of nanoparticles in conducting polymers has further increased the performance of the sensors by enhancing the electrical conductivity and percolation features of the nanomaterials [9]. However, various non-conducting polymers have also been applied to form nanomaterials for sensors [10]. Carbon nanoparticles (graphene, graphene oxide, reduced graphene oxide) and inorganic or metal nanoparticles have been used as effective reinforcements [11,12]. The nanocomposite sensors showed good sensitivity towards the electronic signals as well as chemical, biological, ionic, and gaseous species [13]. The nanocomposite sensors had high molecular recognition sensitivity and response towards various analytes, ions, and molecules [14,15]. The nanoparticle dispersion in the nanocomposite and interaction with the matrix define the final sensing properties [16]. For good dispersion, methods like in situ and solution techniques have been utilized [17]. The use of nanocomposite sensors has been detected in a wide range of technical fields like motion-sensing, gas-sensing, chemical detection, and biomolecule-sensing [18]. Though sensors have been developed using conjugated as well as non-conjugated matrices, conductive polymers revealed noteworthy sensing properties. In particular, conducting polymers combined with carbon nanofillers like graphene-related nanofillers or carbon nanotubes may form a better charge transfer complex due to π - π interactions, leading to high sensing properties. Moreover, polymer nanocomposites with inorganic nanoparticles (metal or metal oxide) have been studied; however, no substantial studies have been reported on these materials. As the sensing mechanism seemed dependent on matrix-nanofiller interactions, the organic nature of polymer/nanocarbon nanocomposites led to more enhanced sensing features than in polymer/inorganic nanocomposite designs. Interactions between inorganic nanoparticles and polymers can be enhanced using functional nanofillers for better dispersal. Thus, poor nanoparticle dispersion and interactions with polymers along with an improper processing technique may lead to fewer interactions with the analyte molecules and inadequate sensing properties. Here, novel design strategies need to be established to overcome the dispersion and fabrication challenges of the sensing nanocomposite towards high selectivity, sensitivity, and response time. The sensing mechanisms in the nanocomposites need to be explored thoroughly, especially for the physisorption or chemisorption of analytes on the nanocomposites.

This state-of-the-art review provides an overview of polymeric nanocomposites in sensing applications, and the design, fabrication, features, and performance of various polymers and derivative nanocomposites reinforced with nanocarbons and inorganic nanoparticles are discussed. Accordingly, nanocomposite materials have been developed and found functional for high-efficiency sensing solicitations. The present article essentially aims to highlight the significance of using multifunctional nanocomposites for sensing applications, covering the design specifications, related physical features, processing methods, technical applications, and the main glitches in this direction. As conceded in this state-of-the-art overview, several polymers have been amalgamated with nanofillers for sensor designs, desired physical features, and sensing performances such as response, sensitivity, selectivity, etc., towards several analyte species. To the best of our knowledge, this review is novel in the literature due to uniqueness of its outline and the literature enclosed, relative to any previous reports in the literature. Moreover, this review is comprehensive when expressing the essential technical as well as viable progresses of nanocomposites in the sensing sector.

2. Design and Features of Multifunctional Nanocomposites Applied for Sensing

2.1. Nanocarbon-Reinforced Nanocomposites

Graphene is a unique carbon nanomaterial with sp^2 hybridized atoms in a one atom-thick nanosheet [19]. It has a two-dimensional structure and a range of exclusive structural and physical properties. Graphene exist in number of modified forms or derivatives such

as graphene oxide, reduced graphene oxide, functional graphene, and functional graphene oxide [20]. To convert graphene to graphene oxide, Hummer's method has been found successful. Similarly, for the conversion of graphene oxide to reduced graphene oxide, various reducing agents have been used. Graphene as well as the derivatives have been found to be efficient regarding significant sensor applications [21,22]. In particular, the formation of conducting polymer- and graphene-derived nanocomposites led to high sensing responses [23–25]. A sensor response can be referred as relative change in resistance [26]. It can be determined as the ratio of sensor resistance in contact with desired analyte and resistance in the absence of analyte. Polyaniline, an important conducting polymer, reinforced with graphene or graphene-based forms, has been used for gas-sensing, ion-sensing, glucose-sensing, and bio-sensing purposes [27–29]. Wu et al. [30] designed the polypropylene, polyaniline, and graphene nanofiller-derived nanocomposite sensor. Facile in situ polymerization and dip coating techniques were applied to form nanomaterials. The designed nanocomposite sensor was used to detect the volatile sulfur compounds and ammonia gases. The fabrication of polypropylene/graphene/polyaniline nanocomposite sensor is given in Figure 1. The in situ method using surfactants was employed to form a nanocomposite sensor. The organization of gas-sensing testing system is also presented. The as prepared nanocomposite film was placed in a test chamber. The controlled amounts of desired gases were injected using mass flow controllers. Figure 2 illustrates the suggested sensing mechanism of nanocomposite film. Sensing performance seemed dependent upon the interface formation in the polypropylene/graphene/polyaniline nanocomposite. Moreover, polyaniline/graphene developed interconnecting network nanostructure within hierarchically porous polypropylene matrix. Thus, several effective conducting pathways have been developed in the matrix. Consequently, reversible doping/de-doping occurred at the interface of polypropylene/graphene/polyaniline nanocomposite to form charge carriers for rapid sensing responses [31]. Interface formation and the superior conductivity of nanocomposite caused a high sensing response towards the ammonia molecules. Figure 3 shows the sensing response towards the detection of volatile sulfur compounds in exhaled garlic breath. The sensor response revealed only around 2% of H₂S gas detected in breath, suggesting a low non-toxic amount of sulfur compounds. The nanocomposite sensor also showed a detection limit of 100 ppb and a fast response of 114 s for the ammonia NH₃ gas-sensing. The response of the polypropylene/graphene/polyaniline nanocomposite sensor was approximately 250% higher than in neat polyaniline sensor. Several combinations of the polyaniline/graphene and the polyaniline/graphene oxide nanocomposites have been utilized for sensing gases and ions [32]. These nanocomposites revealed improved the electrical conductivity and sensing in terms of various gases like hydrogen, methane, methanol, and ions (Zn(II), Pb(II), Cd(II), etc.) [33–35].

Chuiprasert et al. [36] formed polyaniline, poly(*o*-phenylenediamine), and reduced graphene oxide-derived sensors for human health monitoring. The sensor was developed via molecularly imprinting polymer technology onto a glassy carbon electrode. For ciprofloxacin, sensors revealed a good detection limit and sensitivity of 5.28×10^{-11} mol L⁻¹ and 5.78 μ A mol⁻¹ L, correspondingly. The sensing device also had good reproducibility. Zhou et al. [37] fabricated a nanocomposite sensor of polyaniline, natural rubber, and reduced graphene oxide through an in situ method. Graphene oxide was converted to reduced graphene oxide in situ and then the polymerization of aniline led to nanocomposite formation (Figure 4). The in situ method caused the good dispersion of reduced graphene oxide in the polyaniline and natural rubber blended matrices. Homogeneous nanofiller dispersion was supportive for enhancing the sensing performance of nanocomposites. On the other hand, simple blends of polyaniline, natural rubber, and reduced graphene oxide were also prepared without using an in situ technique. The in situ-formed polyaniline/reduced graphene oxide/natural rubber nanocomposite sensor was connected to a blue LED light to observe the chemical sensing features via changes in illumination (Figure 5). The LED light connected to nanocomposite sensor was dipped in toluene. The light faded due to an increase in electrical resistivity. After its removal from the toluene, the LED light

regained illumination and worked for 8 min. Outcomes proposed a commercial level use of polyaniline/reduced graphene oxide/natural rubber nanocomposite sensor for chemical sensing purposes.

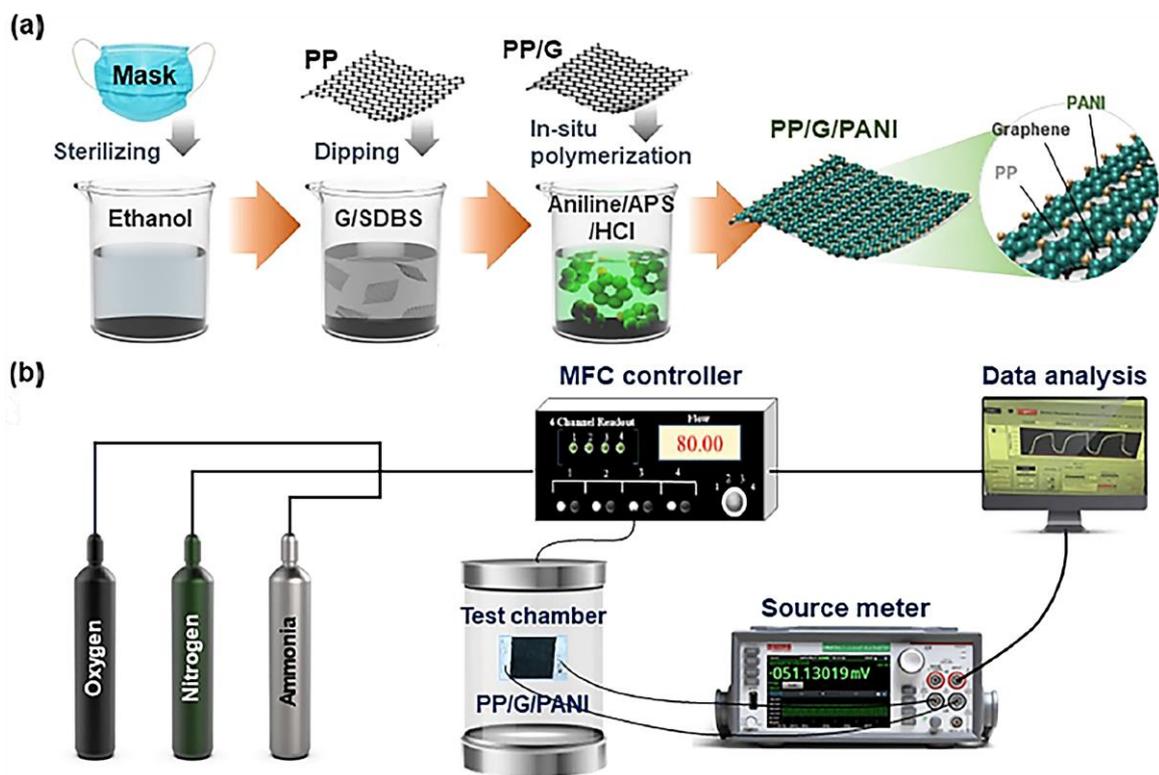


Figure 1. (a) Schematic diagram of the flexible PP/G/PANI hybrid sensor preparation process; and (b) organization of the gas-sensing testing system [30]. PP = polypropylene; PANI = polyaniline; G = graphene; PP/G = polypropylene/graphene; PP/G/PANI = polypropylene/graphene/polyaniline; MFCs = mass flow controllers. Reproduced with permission from ACS.

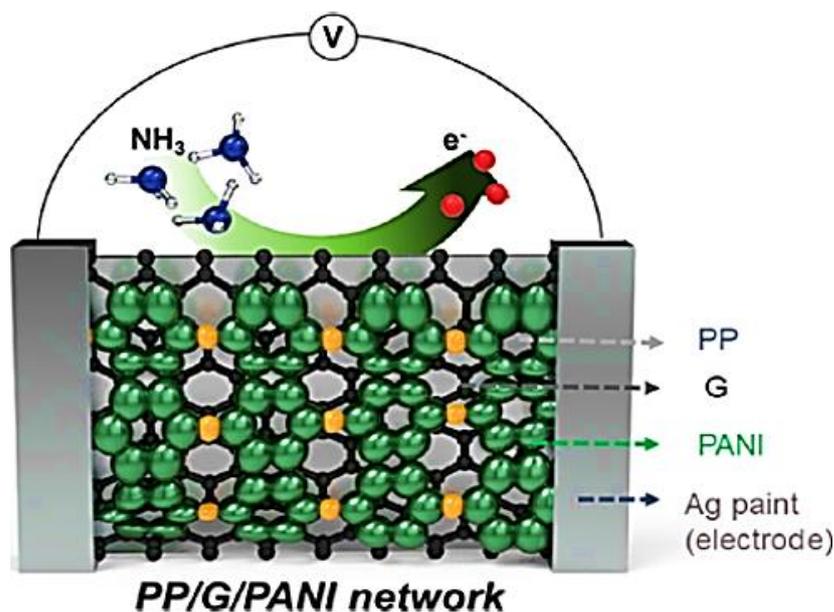


Figure 2. Sensing mechanism of the PP/G/PANI hybrid sensor [30]. PP = polypropylene; PANI = polyaniline; G = graphene; PP/G/PANI = polypropylene/graphene/polyaniline. Reproduced with permission from ACS.

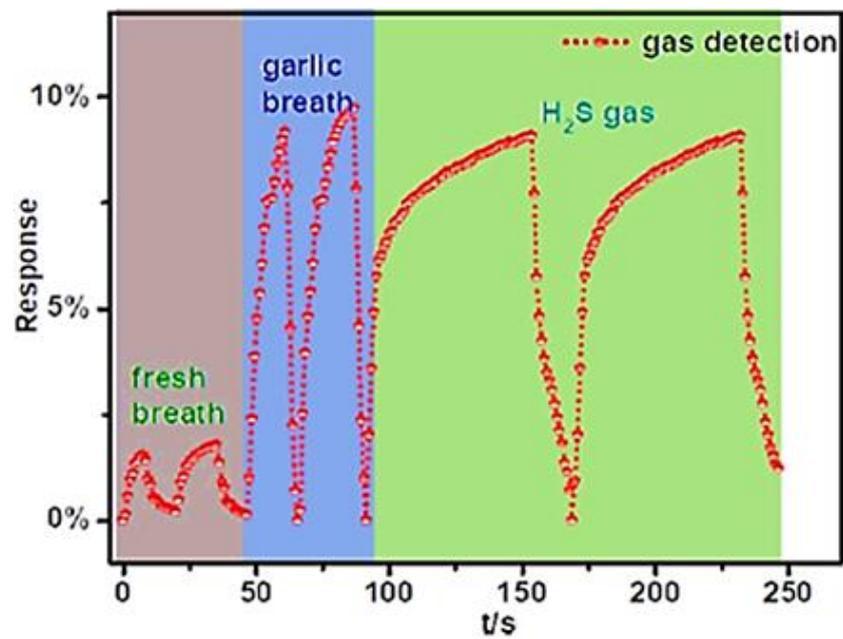


Figure 3. Photograph of the flexible PP/G/PANI sensor for volatile sulfur compounds (VSCs) in exhaled breath and pure H₂S. [30]. PP/G/PANI = polypropylene/graphene/polyaniline; reproduced with permission from ACS.

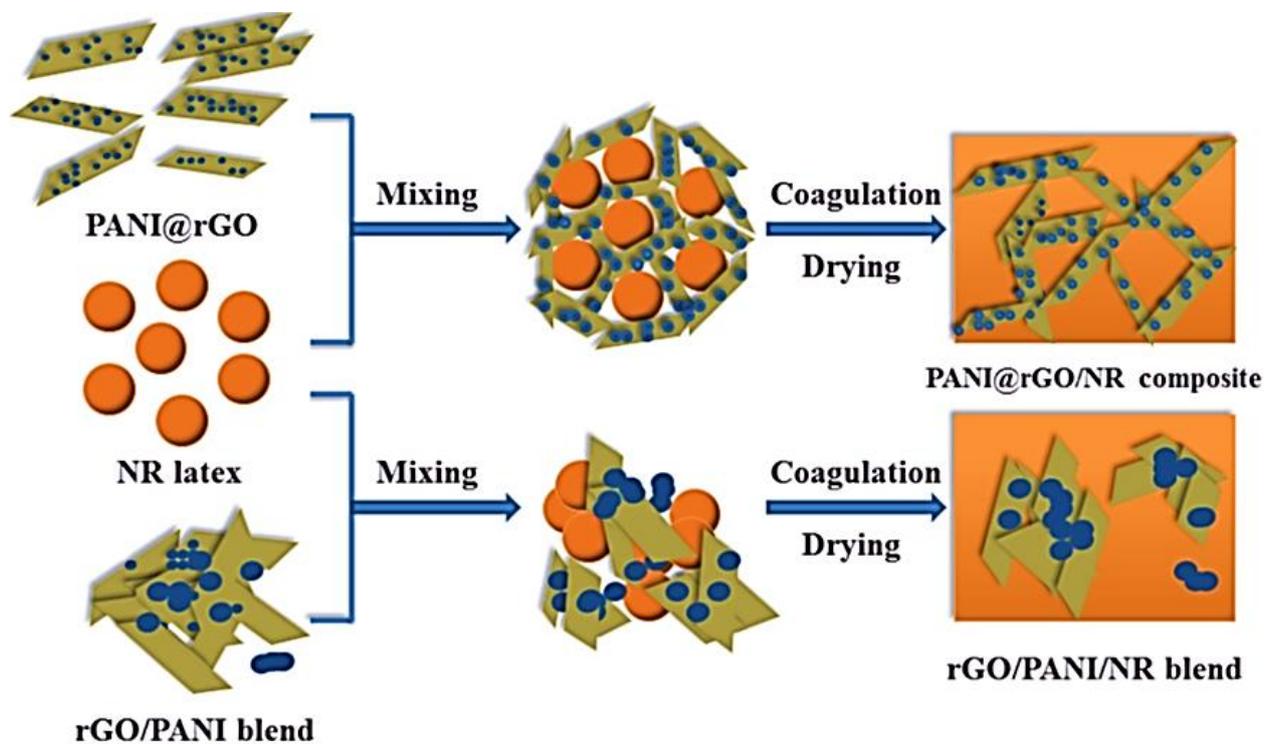


Figure 4. Schematic strategy of fabrication of PANI@rGO/NR composite and rGO/PANI/NR blend [37]. PANI@rGO/NR = polyaniline@reduced graphene oxide and natural rubber. Reproduced with permission from Elsevier.

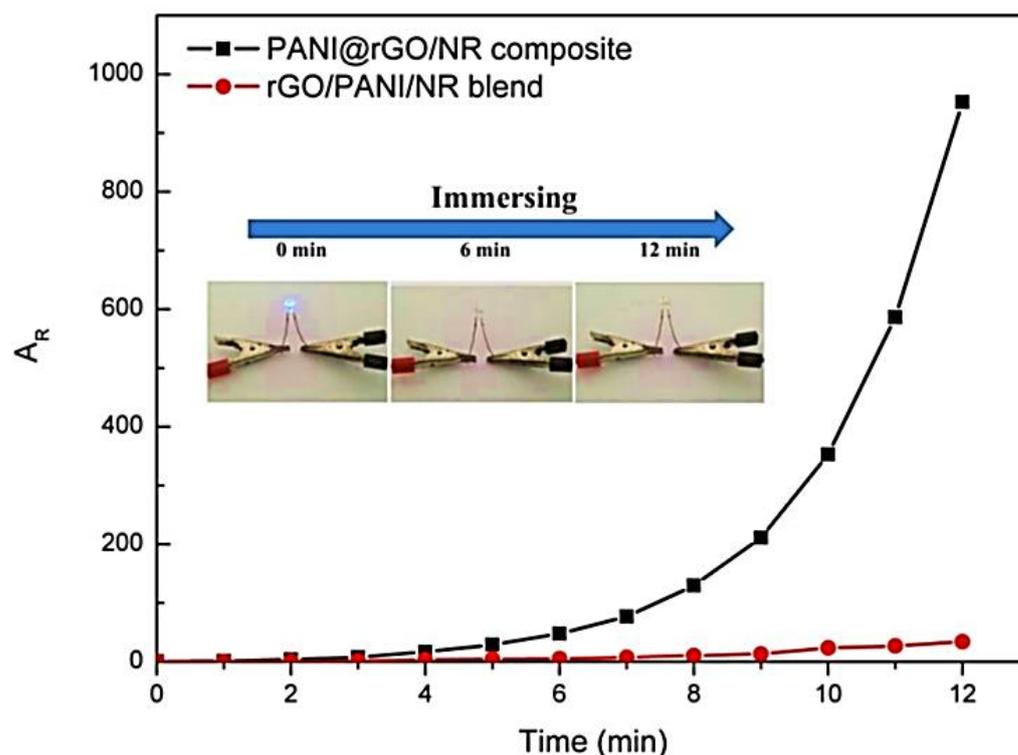


Figure 5. The relative resistance change plots as a function of time and photos of illumination changes during PANI@rGO/NR nanocomposite immersion in toluene (inset) [37]. PANI@rGO/NR = polyaniline@reduced graphene oxide and natural rubber; rGO/PANI/NR = reduced graphene oxide/polyaniline/natural rubber. Reproduced with permission from Elsevier.

Carbon nanotubes are one-dimensional hollow nanotubes of sp^2 hybridized carbon atoms [38,39]. Carbon nanotubes have large surface areas and superior structural and physical features. Their unique nanostructure was found to be responsible for superior sensing behaviors due to capturing ions, chemicals, and gaseous molecules [40]. Carbon nanotubes also possess inert nature and ecological constancy [41,42]. Additionally, carbon nanotubes have fast electron conduction and charge transference properties [43–45]. Sensors based on carbon nanotubes have good sensitivity, rapid responsiveness, and selectivity characteristics [46,47]. In the form of gas sensors, carbon nanotubes reveal good interaction with gas molecules via adsorption or bonding modes [48]. Generally, conducting polymer and carbon nanotube may form donor acceptor sites for electron or charge transference [49–51]. For example, carbon nanotubes have caused changes in electron conductivity upon interaction with hydrogen and other gas molecules [52,53]. Among the conjugated polymers, polyaniline has been greatly studied for its gas-sensing applications. Combinations of polyaniline matrix and single-walled carbon nanotube or multi-walled carbon nanotube have been applied to form efficient sensing devices [54]. Srivastava et al. [55] developed change in resistance sensors for hydrogen gas. Thus, polyaniline, polyaniline/single-walled carbon nanotubes, and polyaniline/multi-walled carbon nanotube nanocomposites were fabricated via a solution technique. Afterwards, the nanomaterials were spin-coated to use as sensors [56]. The spin-coated films were deposited on an indium tin oxide coated glass substrate. Table 1 illustrates the change in the electrical resistance response of these sensors towards hydrogen gas. The resistance change was due to the good gas sensitivity of the sensors. The superior performance was credited to high a surface area and the density of nanocomposite materials.

Table 1. The change in resistance and conforming response of PANI, PANI/SWCNT, and PANI/MWCNT nanocomposite sensors for hydrogen gas [55]. PANI = polyaniline; PANI/SWCNT = polyaniline/single-walled carbon nanotube; PANI/MWCNT = polyaniline/multi-walled carbon nanotube. Reproduced with permission from Elsevier.

Sample	Initial Resistance (Air)	Shift in Resistance (2% H ₂ in Air)	Response (R _g /R ₀)
PANI film	3.10 MΩ	2.05 ± 0.02 MΩ	1.66
PANI/SWCNT	69.2 kΩ	57.2 ± 0.09 kΩ	1.83
PANI/MWCNT	99.1 kΩ	129 ± 0.1 kΩ	2.30

Gao et al. [57] reported efficient motion sensor based on polyurethane/carbon nanotube nanocomposite-derived helical yarn. The designed sensor was low weight, flexible, and wearable, as well as stretchable. Due to the synergistic effects of polyurethane and carbon nanotubes, the nanocomposite developed winding locks in the form of yarns. Figure 6 shows the strain-sensing performance of a polyurethane/carbon nanotube nanocomposite helical yarn sensor. Its performance was evaluated during finger-, arm-, wrist-, and leg-bending movements. The strain sensor exhibited resistances of 2–5 kΩ, during movements. The linearity in resistance and repeatability were observed in the real time movement curves. The sensor had a recoverability of 900% and a tensile elongation of ~1700%. By varying nanotube contents, sensor response rate was also improved. Thus, these strain sensors were suggested for application in low-price, stretchable, and wearable electronics, such as human body sensors.

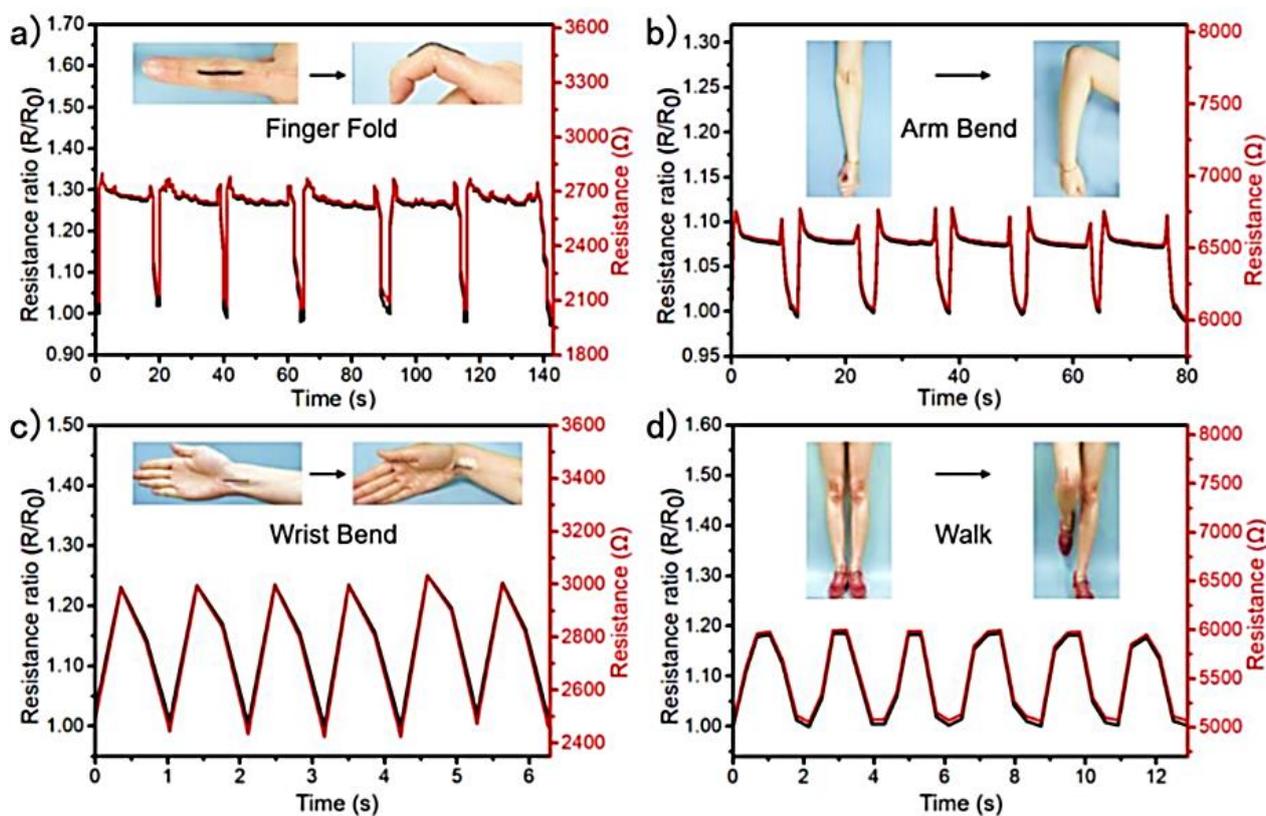


Figure 6. Applications of polyurethane/carbon nanotube helical yarn as a strain sensor to monitor resistance changes during (a) finger-folding, (b) arm-bending, (c) wrist-bending, and (d) walking. Fast signal response and stable resistance repeatability for various human motion was observed [57]. Reproduced with permission from ACS.

2.2. Inorganic Nanoparticle-Reinforced Nanocomposites

In addition to carbon nanoparticles, inorganic nanoparticles have inimitable sensing features when reinforced in the conducting polymer matrices [58]. The unfilled conducting polymers reveal good electron transference properties and low sensitivity properties due to weak interactions with analyte species [59]. On the other hand, the inclusion of inorganic nanoparticles in conductive matrices significantly improved the sensing characteristics of conducting polymer/inorganic nanoparticle nanocomposites [60]. Generally, the conducting polymers are filled with metal or metal oxide nanoparticles to fabricate the sensors [61]. In nanocomposites, the desired analyte molecules efficiently diffuse the materials, leading to an active sensing response [62]. Nevertheless, designing conducting polymer/inorganic or metal nanoparticle nanocomposites for sensing purposes face several challenges [63]. With polyaniline matrix, tin oxide, and zinc oxide nanofillers have revealed efficient sensing responses [64,65]. Such nanomaterials have been specifically found operative to sense ammonia gas [66]. Moreover, the mixing of copper nanoparticles with polyaniline matrix reveal a good sensing performance to detect chloroform [67,68]. The sensing properties of polyaniline-derived nanocomposites were reliant on nanofiller dispersion and contents. In addition, polyaniline and polypyrrole were also utilized to fabricate nanocomposite sensor with various inorganic nanoparticles [69]. Consequently, polypyrrole/tin oxide- and polypyrrole/zinc oxide-derived nanomaterial sensors have been reported [70–72]. These sensors have also been utilized for ammonia recognition. Similarly, the alumina nanoparticles with polypyrrole matrix have yielded good ammonia-sensing features [73–75]. Henceforth, the addition of inorganic nanofillers has been found to improve the electron conduction and the gas-, ion-, or chemical-sensing performance of the nanomaterials [76].

3. Technical Applications of Multifunctional Nanocomposites towards Various Sensors

3.1. Strain-Sensing

In strain-sensing applications, the nanomaterials must have good electrical or optical responses. Thus, innumerable polymer nanocomposites have been used in strain sensors. Most importantly, polyaniline [77,78], polypyrrole [79], poly(3,4-ethylenedioxythiophene) [80], and other conducting polymers have been used to develop strain sensors. For this purpose, these polymers have been combined with nanocarbons like graphene [81,82], carbon nanotube [83,84], carbon black [85,86], and other nanoparticles [87,88]. These nanoparticles have been found to develop conducting pathways in polymers to create a percolation threshold [89,90]. The percolation threshold is a concept adopted from percolation theory, which designates the development of long-range connectivity in random systems like nanocomposites [91]. Here, the percolation threshold conforms to the nanofiller content, above which the electrical conductivity properties of nanocomposite increase in an exponential way, due to the development of interlinked pathways in the matrix [92]. Competent electron transportation and percolation features resulted in the good strain-sensing properties of nanocomposites [93–95]. Subsequently, nanocomposite sensors may easily monitor the applied strain effects [96]. Investigations on electron conduction and microstructure properties have been found essential to unfold the strain-sensing effects of nanocomposites [97]. Moreover, nanocomposite sensors have excellent durability, stretchability, and sensitivity properties [98]. Strain sensors had beneficial applications in human health diagnosis [99], motion detection [100], and facile wearable electronics [101,102]. Non-conducting thermoplastic elastomers (poly(dimethylsiloxane), polyurethane, poly(styrene-butadiene-styrene), natural rubber, isoprene rubber, and block copolymers) have also been employed to develop strain sensors [103]. In some cases, blends of conjugated and non-conjugated polymers have been used to form nanocomposite strain sensors. Ding et al. [104] developed thermoplastic polyurethane, hydroxyethyl cotton cellulose nanofibers, and carbon nanotube-derived nanocomposite as strain sensors. Figure 7 shows the formation route of the nanocomposite [105,106]. For carbon nanotube and hydroxyethyl cotton cellulose nanofiber nanofillers,

a freeze-drying method was used [107,108], whereas the solution procedure and curing approaches were applied to form nanocomposites. Nanofiller functionalities have hydrogens that bond with polyurethane matrix and a strong interface. Consequently, thermoplastic polyurethane/hydroxyethyl cotton cellulose nanofibers/carbon nanotube nanocomposites have been effectively used as a wearable electronic device. Human motion-sensing performance was investigated in the form of finger movements (Figure 8). The 90° finger bending motion was studied. The finger movements produced periodic sensing signals at an adjustable strain [109,110]. The nanocomposite sensor had a shape fixity ratio and a shape recovery of 49.7% and of 76.6%, respectively. Efficient sensors have been found to be effective for next generation smart devices.

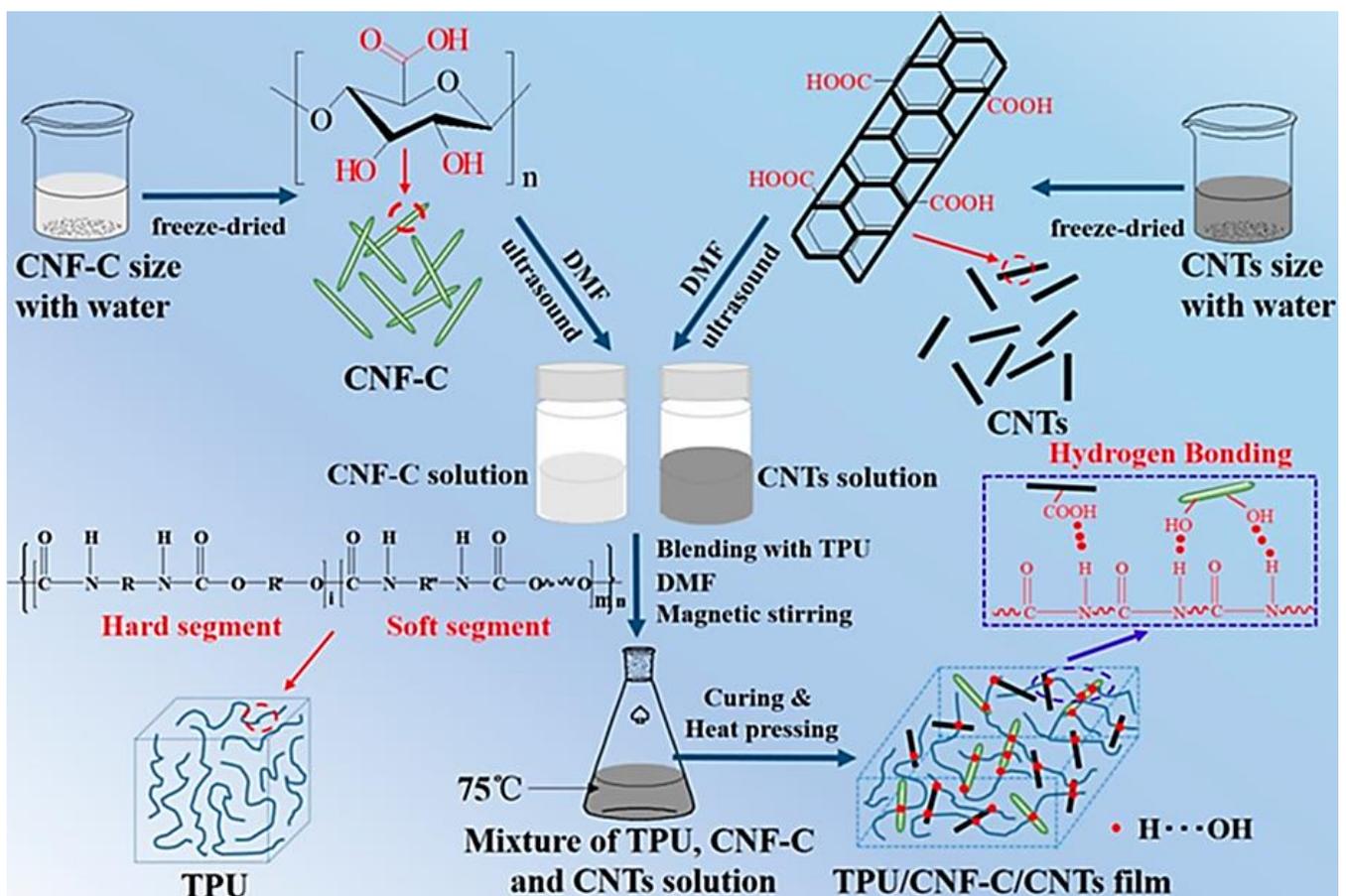


Figure 7. Schematic diagram of the hydrogen bonding interaction mechanism among the molecules of TPU, CNF-C, and CNTs and the film formation of TPU/CNF-C/CNTs [111]. TPU = thermoplastic polyurethane; CNF-C = hydroxyethyl cotton cellulose nanofibers; CNTs = carbon nanotubes; TPU/CNF-C/CNTs = thermoplastic polyurethane/hydroxyethyl cotton cellulose nanofibers/carbon nanotubes; DMF = dimethyl formamide. Reproduced with permission from MDPI.

3.2. Gas-Sensing

For gas sensors, polymer- and nanocarbon-derived nanocomposites revealed tunable electron conductivity, strength, durability, and chemical-sensitive features [112]. Interactions of graphene or derivatives, carbon nanotube, fullerenes, etc. with polymers developed extended π -systems in order to contribute towards efficient electron conduction. In addition, the nanocomposites of polymers and the range of metal oxides (tin oxide, silver oxide, zinc oxide, titania, and many more) have been reported [113–115]. Consequently, high performance nanocomposites have good sensitivity and selectivity for toxic gases like the oxides of carbon, sulfur, nitrogen, and organic vapors [116]. Essential conjugated polymers like polyaniline, polypyrrole, and polythiophene reveal NO_x detection prop-

erties [117]. In addition, conductive nanocomposites reinforced with nanoparticles have sensing properties for halogens, methane, nitrogen or sulfur oxide, and other gaseous species [118,119]. For gas-sensing, polyaniline nanocomposites can sense CO and NO_x species [120]. The polypyrrole nanocomposite-derived gas sensors can also efficiently detect NO_x molecules [121]. Likewise, polythiophene nanocomposite-derived sensors may sense hydrazine gas [122]. All these sensors have high sensitivity and reproducibility features. For example, the nanomaterials based on polyaniline/tin oxide [123], polypyrrole/tin oxide [124] polyhexylthiophene/tin oxide [125], poly(ethylene dioxythiophene)/tin oxide [126], etc., have been studied in order to sense hydrocarbons and NO_x gases. All these sensing materials revealed high sensitivity and selectivity. Deshpande and co-workers [127] produced polyaniline/tin oxide nanocomposite-based gas sensors. The nanomaterial was formed via the in situ polymerization of aniline in the presence of ammonium peroxydisulfate oxidizing agent and tin oxide nanoparticles. Nanocomposites were tested to sense ammonia gas. Figure 9 demonstrates that the plots of sensitivity versus concentration for neat polyaniline, tin oxide nanoparticles, and nanocomposites. At 300 ppm of ammonia gas, the polyaniline/tin oxide nanocomposite sensor showed a rapid response time of 12–15 s and recovery times of around 80 s. The good performance of gas sensors was observed due to the formation of conducting channels via the interactions of matrixes and nanofillers in nanocomposites, relative to the individual components.

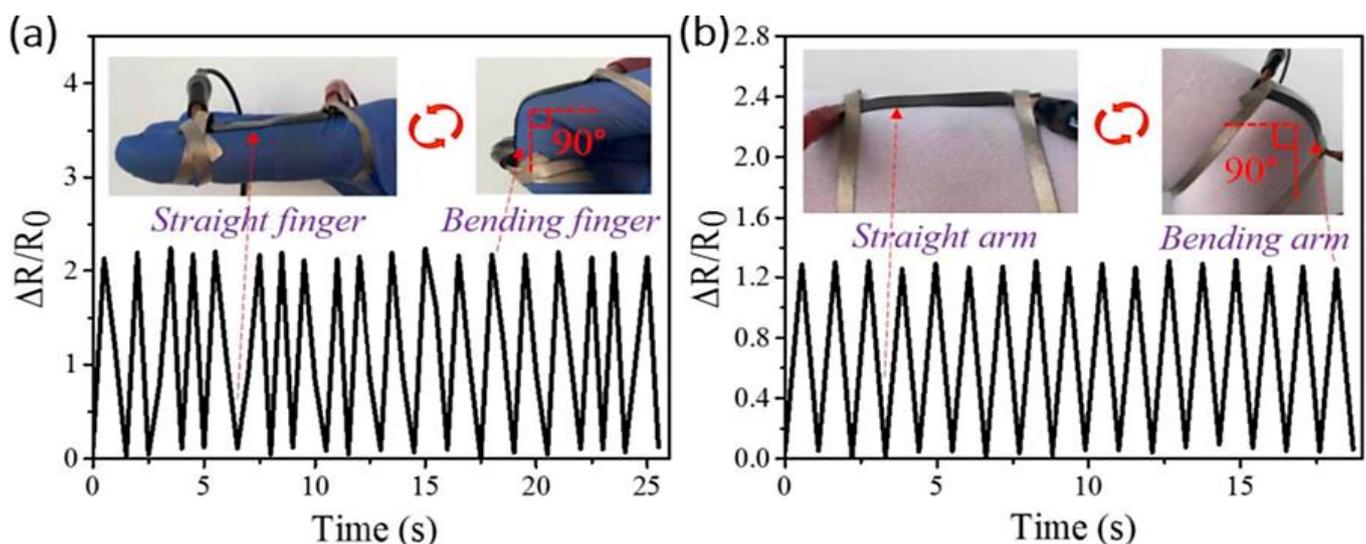


Figure 8. Human motion monitoring using the as-fabricated sensor: (a) relative resistance variation ($\Delta R/R_0$) versus time for blending and release of the index finger; inset: snapshots of one bending cycle. (b) $\Delta R/R_0$ for TPU/CNF-C/CNTs strain-sensors according to bending motions; inset: one bending cycle of arm, the bending motion degree of the arm at angle of 90° [111]. TPU/CNF-C/CNTs = thermoplastic polyurethane/hydroxyethyl cotton cellulose nanofibers/carbon nanotubes. Reproduced with permission from MDPI.

Table 2 shows that certain essential polymers and nanocarbon or inorganic nanoparticles are derived from nanocomposite-based sensors. Polymeric nanocomposites revealed remarkable gas-sensing features with graphene, graphene oxide, carbon nanotubes, and inorganic oxides due to interactions and charge transfer complex development. Nanocomposites may act as electron donor–acceptor nanostructures for the analyte molecules.

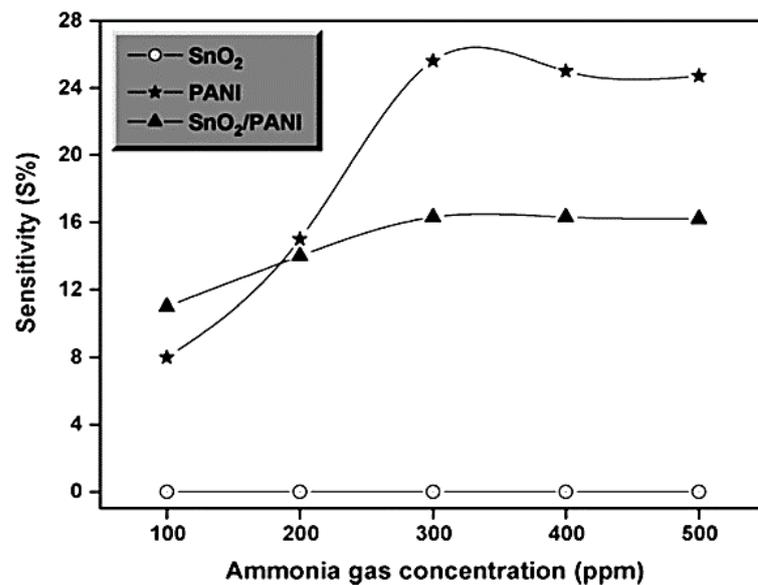


Figure 9. Sensitivity versus concentration plots for tin oxide, polyaniline, and polyaniline/tin oxide nanocomposites [127]. Reproduced with permission from Elsevier.

Table 2. Specifications of some important polymer nanocomposite-based sensors.

Conjugated Polymer	Nanofiller	Processing	Property/Application	Ref
Polyaniline	Carbon nanotube	Spin coating method	Hydrogen gas-sensing	[55]
Polyaniline	Carbon nanotube	Interfacial technique	Ammonia-sensing	[128]
Polypyrrole	Carbon nanotube	Spin coating	Ammonia-sensing	[129]
Polypyrrole	Carbon nanotube	In situ and spin coating methods	Ammonia sensor	[130]
Polyaniline	Graphene	Interfacial technique	H ₂ O ₂ -sensing	[131]
Polyaniline	Graphene oxide	In situ method	Methanol sensitivity; electrical conductivity 241 Sm ⁻¹	[132]
Polyaniline	Graphene	Layer-by-layer technique	π - π conjugation; high methane sensitivity	[133]
Polythiophene	Reduced graphene oxide	In situ method	Humidity sensor	[134]
Polyaniline	ZnO-SnO ₂	In situ chemical method	Ammonia-sensing	[135]
Polyaniline	Silver nanoparticle	In situ technique	Ammonia-sensing; sensitivity $\sim 12.5 \mu\text{AmM}^{-1} \text{cm}^{-2}$; response time 10 s	[136]
Polypyrrole	ZnO-TiO ₂	In situ method	Ammonia-sensing	[137]
Polypyrrole	SnO ₂ -ZnO	In situ process	Ammonia-sensing	[73]
Polypyrrole	Ag-ZnO	In situ process	Ammonia-sensing	[76]

3.3. Bio-Sensing or Chemical-Sensing

To monitor the biological or chemical species, various nanocomposite sensors have been developed [138]. For this purpose, metal nanoparticle-derived nanomaterials exhibited good biomolecular sensing [139]. In particular, metal nanoparticle nanocomposite sensors have been applied to assess biological and chemical toxins with high sensitivity and selectivity [140]. Thus, these sensors may also assist in monitoring the chemical processes occurring in living systems. Relative to traditional sensors, nanocomposite sensors

revealed sensing properties that were several times better in chemical process monitoring [141]. Segets et al. [142] applied zinc oxide nanoparticles of 10 nm to monitor in situ chemical growth reactions in the biological systems. Consequently, zinc oxide nanoparticles are frequently applied to biologically sense proteins, deoxyribonucleic acid (DNA), ribonucleic acid (RNA), etc., in order to detect biological ailments [143–145]. Gold nanoparticles have also been used in active biosensors [146,147]. For instance, DNA and RNA sequence recognition has been performed via the absorption mechanism and the use of gold nanoparticles and related nanocomposites [148,149]. Furthermore, biological species can also be detected through the electrostatic interactions between the oligonucleotide and nanocomposites [150–152]. Li et al. [153] studied the biodegradability and enzyme-sensing features of the amyloid fibrils (natural protein) and graphene-derived nanocomposite sensors. Nanocomposites exhibited high conductivity and degradation behavior and can thus be employed as efficient biosensors. Figure 10 demonstrates the fabrication of the amyloid fibrils/graphene nanocomposites. The amyloid fibrils were prepared at a high temperature of >80 °C. The amyloid fibrils were then coated with graphene dispersion. Figure 11 presents the enzyme-sensing properties of the amyloid fibrils/graphene nanocomposite. The pepsin enzymatic activity was evaluated using the biosensor. The pepsin activity of biosensor was increased with time due to the perfectly formed natural protein–graphene sensor. Subsequently, numerous sensors have been applied for bio- or chemical-sensing applications [154]. Nanocomposite-derived biosensors have the capability of sensing the biological species through electrical, optical, or magnetic interactions [155]. Such biosensors have high durability, non-toxicity, sensitivity, and selectivity features regarding the detection of multiple biological or chemical molecules or ions [156].

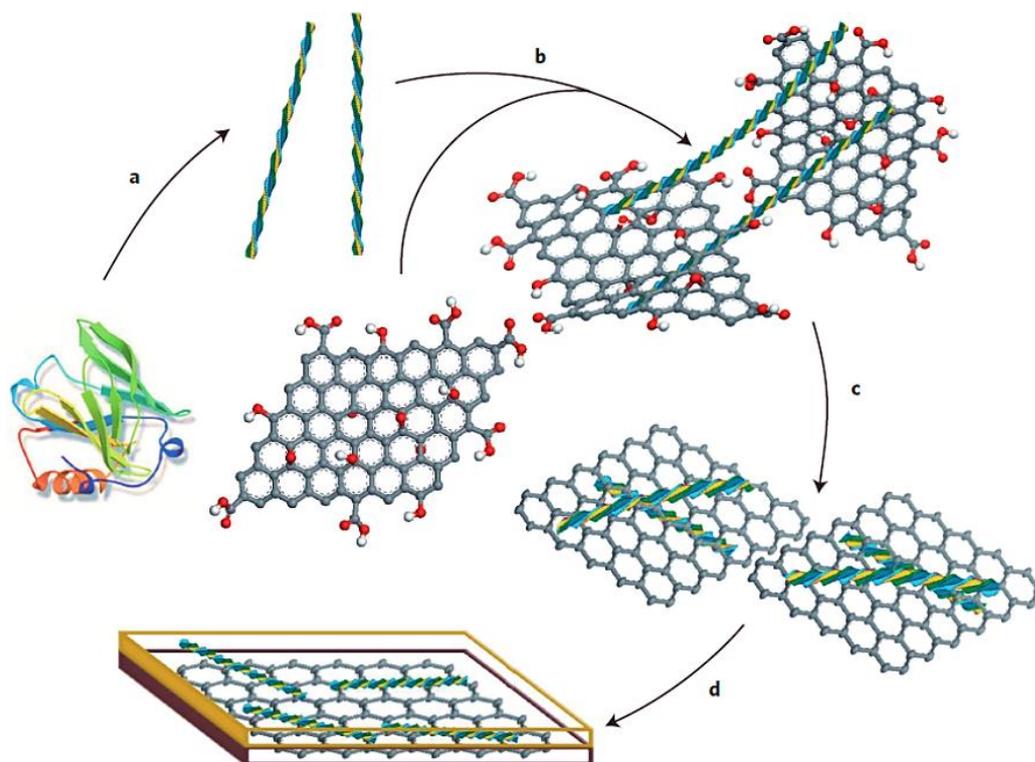


Figure 10. Graphic representation of the formation of free-standing films of amyloid fibrils and graphene-derived nanocomposites: (a) the supramolecular self-assembly of b-lactoglobulin into amyloid fibrils at pH 2 and 90 °C; (b) the electrostatic aggregation of amyloid fibrils and graphene oxide; vigorous stirring is required to prevent sedimentation in the system; (c) preferential binding of broken amyloid fibrils on graphene surfaces during the reduction of graphene oxide at 80 °C under vigorous stirring; and (d) the layered organization of amyloid fibrils and graphene nanosheets into hybrid nanocomposites using vacuum filtration [153]. Reproduced with permission from *Nature*.

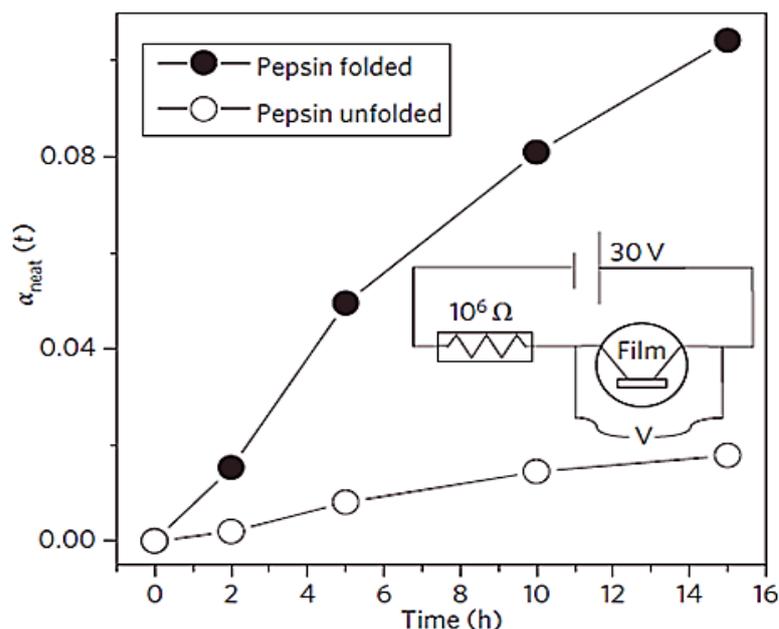


Figure 11. Evolution of the pristine cumulative pepsin activity, $\alpha_{neat}(t)$, measured on the 1:8 hybrid film for 1 wt.% pepsin in a native (folded) and denatured (unfolded) state. Inset: the schematic of the biosensor designed to probe enzymatic activity [153]. Reproduced with permission from *Nature*.

4. Prospects and Conclusions

High performance nanomaterials have been found to be remarkable contenders in the field of sensors, such as strain sensors, gas sensors, biosensors, or chemical sensors [157]. Advancements in nanocomposite sensor technology have filled the gap between the initial design and its real-world application. In nanocomposite sensors, nanofiller type, nanofiller concentrations, polymer type, nanoparticle dispersion in matrix, and matrix–nanofiller interactions contribute greatly to improving the sensing performance of the nanomaterials. Consequently, various types of sensors have been developed to sense the strain-related effects, electrical or optical changes, gases, ions, toxic gases, chemical compounds, proteins, DNA, RNA, and related biomolecules. Hence, nanocomposite sensors have found practical utilization in wearable electronics, and practical gas-sensing devices and biosensors [158]. Accordingly, nanocomposite sensors displayed high durability, sensitivity, selectivity, and responsiveness in high-tech applications. In particular, nanocomposite sensors have been used in strain sensors, gas or ion sensors, biosensors, and chemical sensors.

Despite the research carried out so far, future efforts are required to design novel nanocomposite sensors of conductive or non-conductive polymers and various nanofillers [159]. Facile ways must be adopted to further enhance nanoparticle scattering in polymers to attain the desired sensing performance. Moreover, the mechanism of sensing via interactions between the nanocomposite material and analytes must be comprehensively studied [160]. The in-depth explorations of sensing mechanism and phenomenon will not only help to interpret the detection process but also assist in monitoring the sensor performance. As numerous sensing systems explained in this article, the mechanism interpretation can better help to fabricate and regulate the sensing behavior of the polymers/carbon nanoparticles and polymer/inorganic nanofiller-derived nanocomposites. The future relies on using nanocomposite sensors in smart electronic and microelectronic devices [161]. Although nanocomposite sensors have countless advantages, their complete utilization in the future devices and systems still faces several technical challenges. Using shape memory materials could lead to remarkable advancements in the field of nanocomposite sensors. Shape memory nanocomposites can be very useful for creating smart flexible electronics. Applying three-dimensional or four-dimensional printing techniques could be advantageous for fabricating smart sensors [162]. Here, quite a lot of new design variations need to be

investigated in order to apply nanocomposite sensors to electronics, microelectronics, and other device applications [163]. Further efforts on nanoparticle dispersal and compatibility with polymers are of prime importance in order to create high performance nanomaterials.

Briefly, this comprehensive overview outlines the design and critical features of various types of nanocomposite sensors. Here, conjugated as well as non-conductive polymers were filled with carbon, inorganic, or metal nanoparticles to form high efficiency sensors. The exploration of various combinations of polymer matrices and nanoparticles revealed unique characteristics for a myriad of practical sensing applications, such as strain-sensing, gas-sensing, bio-sensing, or chemical-sensing. This review article can certainly guide scientists in their pursuit of progress in the field of technical nanocomposite sensors.

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