

Review Article

A Review on Polyaniline: Synthesis, Properties, Nanocomposites, and Electrochemical Applications

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The development in the use of polyaniline (PANI) in advanced studies makes us draw attention to the presented research and combine it into one study like this one. The unique composition of PANI qualifies it for use in electrochemical applications in addition to many other applications whose use depends on its mechanical properties. Based on this, it is necessary to limit the reactions that produce PANI and the cheapest cost, and then limit the current uses in the formation of nanocomposites with metals, their oxides, and/or carbon nanocomposites in order to determine what is missing from them and work on it again to expand its chemistry. The development in the use of PANI in advanced studies makes us draw attention to the research presented on PANI and combine it into one study. One of the very important things that made PANI possess a very huge research revolution are preparation in a variety of ways, easy and inexpensive, from which a daily product can be obtained with very high purity, as well as its distinctive properties that made it the focus of researchers in various scientific departments. The unique structure of PANI, which is easy to prepare in its pure form or with various chemical compounds including metals, metal oxides, and carbon nanomaterials (such as carbon nanotubes, graphene, graphene oxide, and reduced graphene oxide), qualifies it for use in electrochemical applications. The various studies reviewed showed that PANI gave good results in the applications of super capacitors. In some of the studies mentioned later, it gave a specific capacitance of 503 F/g, cycle stability 85% at 10,000 cycles, energy density 8.88 kW/kg, and power density 96 W h/kg. It was also noted that these values improved significantly when using PANI with its nanocomposites. Because of its good electrical conductivity and the possibility of preparing it with a high surface area with nanostructures in the form of nanowires, nanofibers, and nanotubes, PANI was used as a gas sensor. We have noticed, through the studies conducted in this field, that the properties of PANI as a basic material in gas sensors are greatly improved when it is prepared in the form of PANI nanocomposites, as explained in detail later. From this review, we tried with great effort to shed light on this attractive polymer in terms of its different preparation methods, its distinctive properties, its nanocomposites, and the type of polymerization used for each nanocomposites, as well as its applications in its pure form or with its nanocomposites in the supercapacitor and gas sensor applications.

1. Introduction

During the past few decades, nanomaterials, polymeric materials, and hybrid materials [1–6] have aroused a distinguished

interest in the world of scientific research due to their distinctive chemical and physical properties and their various applications in photonics, electronics, sensors, conductive materials, biotechnology, medicine, and pollution control

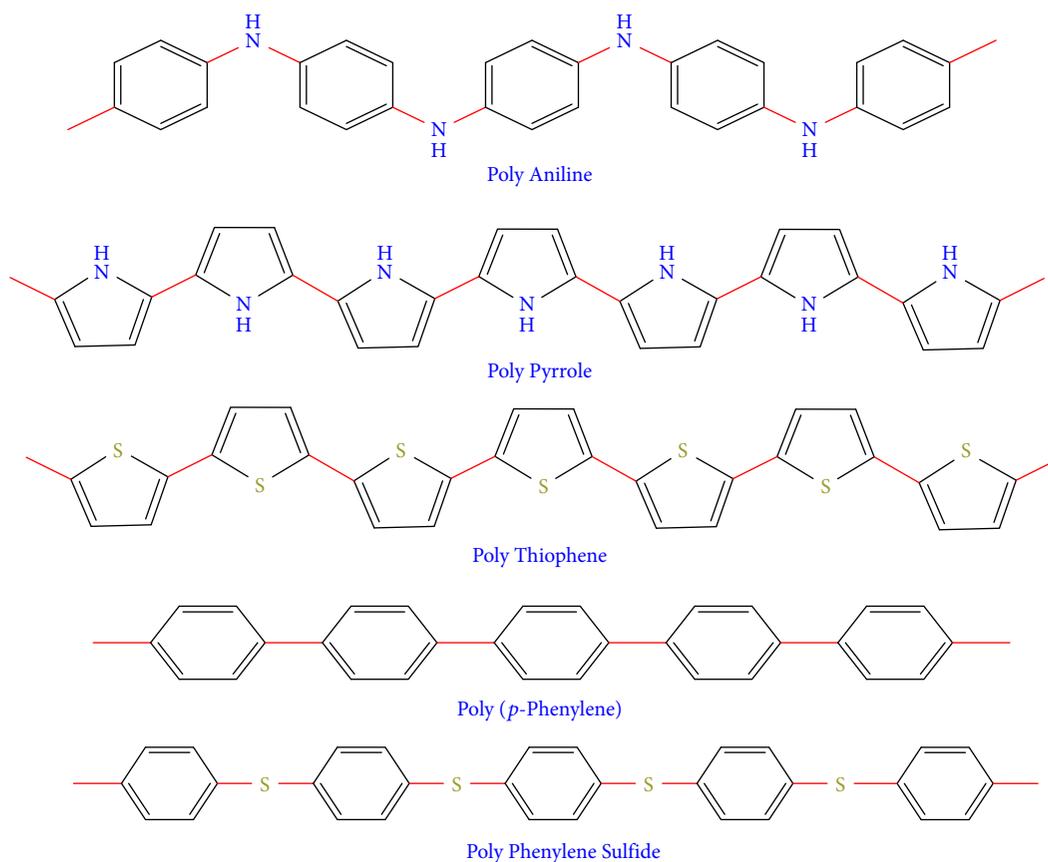


FIGURE 1: Structure of some conductive polymers.

[7–15]. Conductive polymers are among the important classes of polymers that have become popular in recent decades because of their high efficiency, which made them distinguished in the field of electrical conductivity and energy storage [16–18]. Polyaniline (PANI) and its derivatives, polypyrrole, polythiophene, poly(para-phenylene), and polyphenylene sulfide (Figure 1), are among the most widely used classes of conductive polymers in recent times because of their good electrical properties that can be controlled, their good environmental stability, their cheapness, and their easy preparedness [19–21].

2. Polyaniline

PANI is the most important of these polymers mentioned, which has been used in research as an advanced polymeric material during the last decade, because it is a polymer rich in electrons and has good electrical conductivity, as well as it has a good ability to modify and processing [22]. PANI has been used in many applications including solar cells [23], lithium batteries [24], supercapacitors [25], fuel cells [26], flexible electrodes [27], corrosion-resistant coatings [28], water pollutant removal [29], screen printing [30], and sensors [31]. MacDiarmid (1997) suggested different kinds of basic PANI. He proposed three idealized oxidation state structures including completely reduced leucoemeraldine base (LEB), semi-oxidized emeraldine base (EB), and fully

oxidized pernigraniline base (PAB; Figure 2). The EB is the greatest broadly studied PANI type [32, 33].

Different forms of PANI display various colors like leucoemeraldine structure of PANI is colorless or white in color, emeraldine type is blue in base structure and green in salt form, and pernigraniline structure is blue to violet color. Using protonic acids and oxidative doping procedures, EB can be converted into a conducting state (Figure 3), but this process cannot be performed for LEB and PAB [34–36].

The color change associated with PANI in different oxidation states can be used in sensors and electrochromic devices [37]. The best method for making a PANI sensor is arguably to take advantage of the dramatic conductivity changes between the different oxidation states or doping levels [38].

3. Synthesis of PANI

PANI is considered one of the attractive electrical conductive polymers, because it can be easily converted between base and salt forms easily by adding base (OH^-) or acid (H^+). The properties of reduction and oxidation, good electrical conductivity, easy method of preparation, and modification in addition to its environmental stability made PANI one of the most conductive polymeric materials that the researchers dealt with in their work. In the past, the

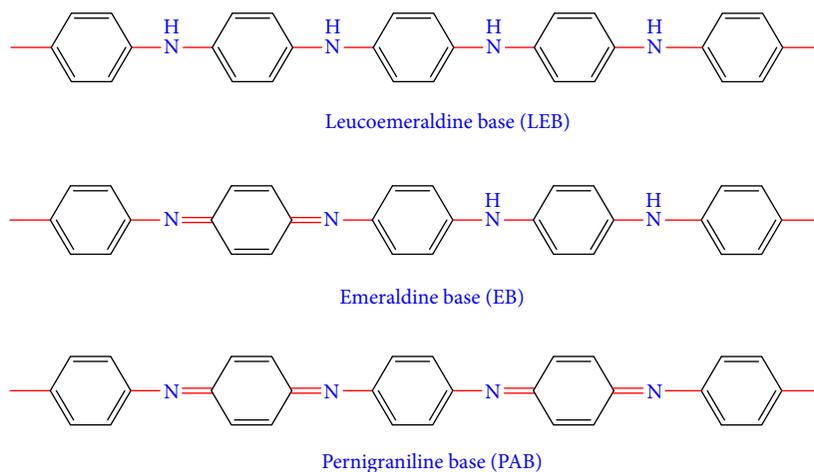


FIGURE 2: The chemical structures of different forms of polyaniline.

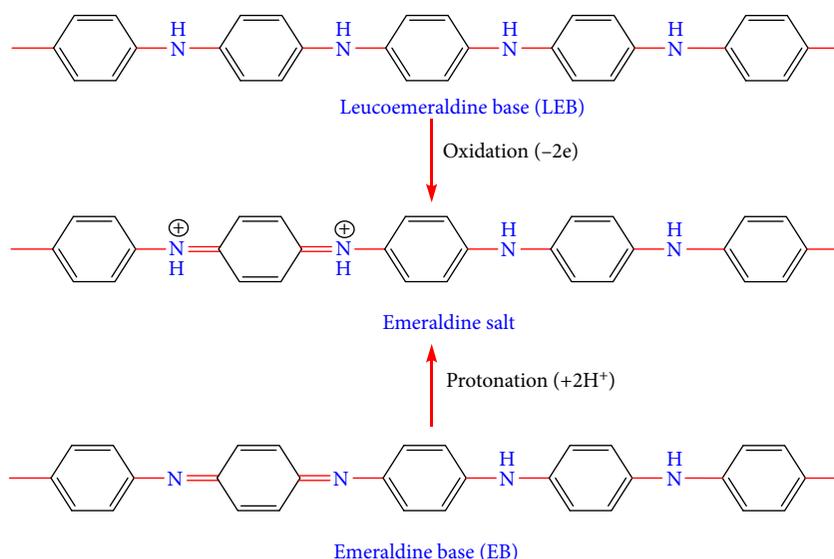


FIGURE 3: Formation of emeraldine salt from leucoemeraldine structure by oxidation and from emeraldine base by protonation.

oxidative polymerization of aniline monomers in an acidic medium was used to prepare PANI, but with the development of work and research on PANI, different methods and techniques were used to prepare it, including [39]:

1. Electrochemical polymerization.
2. Chemical polymerization.
3. Vapor-phase polymerization (VPP).
3. Photochemically initiated polymerization.
4. Enzyme-catalyzed polymerization.
5. Polymerization employing electron acceptors [39].

3.1. Electrochemical Polymerization. The electrochemical method for preparing conductive polymers plays an important role in the preparation, because in many applications, it is required to prepare polymers in the form of a thin film with a wide surface area. When conductive polymers are prepared using the chemical method, it is difficult to follow the electrochemical process of the prepared polymers, because the study of electrical and optical properties depends mainly on these thin layers of polymer deposited

on the electrode. This method of preparing conductive polymers is similar to the electrodeposition method used in metals. Electrodynamics and galvanic techniques were used to prepare PANI as electrochemical methods. The electrochemical method used in the preparation includes many advantages over the chemical method, which is cheap and easy to operate, giving a very pure and homogeneous polymer deposited on the electrode; in addition, the process takes place in a simple chamber cell containing a power source, an electrode, and an electrolyte solution [40, 41].

The electro-polymerization method to prepare PANI follows the next steps: (1) formation of a positive free radical (cationic free radical) of aniline monomers by oxidation at the anode, (2) the combination of the structures formed in the first step to form dimers through the process of removing protons and rearranging electrons in the aromatic rings, (3) the growth of these formed dimers and the formation of new, larger structures, (4) the last step is the spontaneous activation of the polymeric chain formed by the acid present in the solution to obtain the resulting denatured PANI, and this

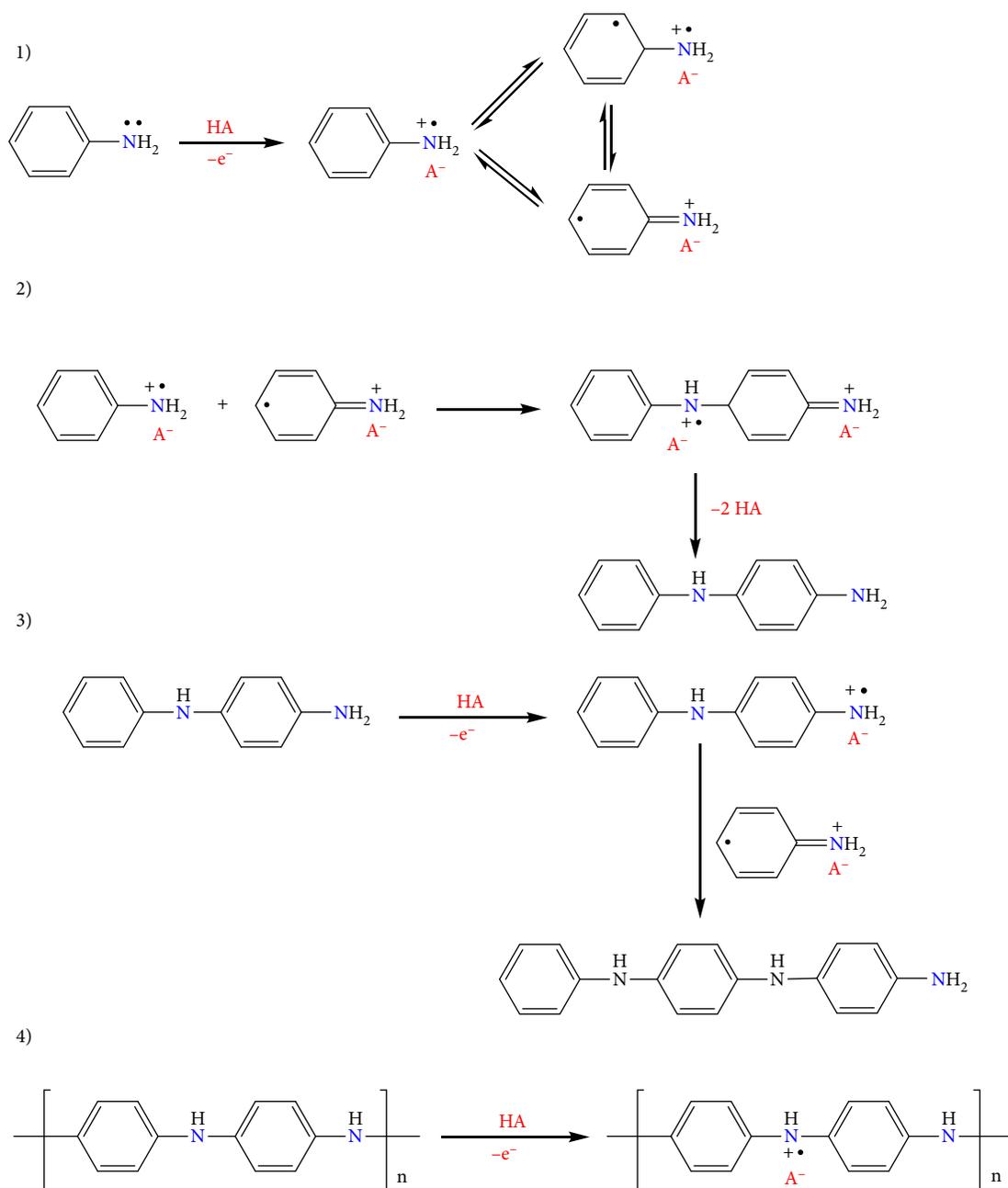


FIGURE 4: Proposal mechanism to form PANI using electrochemical polymerization.

is the proposed mechanism for the electro-polymerization of PANI (Figure 4) [41, 42].

3.2. Chemical Polymerization. The chemical method (oxidative polymerization) for the preparation of polymers is characterized as a simple and cheap method capable of preparing large quantities of polymers within a short time. Until now, it is the common method used by many companies to prepare PANI [43].

In this method, the oxidizing force that initiates the polymerization process is provided by adding a chemical oxidizing agent to the solution. Ammonium persulfate ((NH₄)₂S₂O₈), sodium vanadate (NaVO₃), cerium sulfate (Ce(SO₄)₂), hydro-

gen peroxide (H₂O₂), potassium iodate (KIO₃), potassium dichromate (K₂Cr₂O₇), and others were used as oxidizing agents during the polymerization process [44]. In addition, ammonium persulfate ((NH₄)₂S₂O₈) is one of the most widely used oxidizing materials in this method. Usually, ammonium persulfate is used in the polymerization of aniline in an acidic medium (pH ≤ 3), in order to be suitable for dissolving the aniline, to stimulate the initiation of the polymerization process, and to avoid the formation of unwanted by-products. The usually used molar ratio between persulfate and aniline (APS/Aniline) is less than 1.2 [45].

The mechanism of polymerization of aniline by ammonium persulfate occurs in its initial stages through a

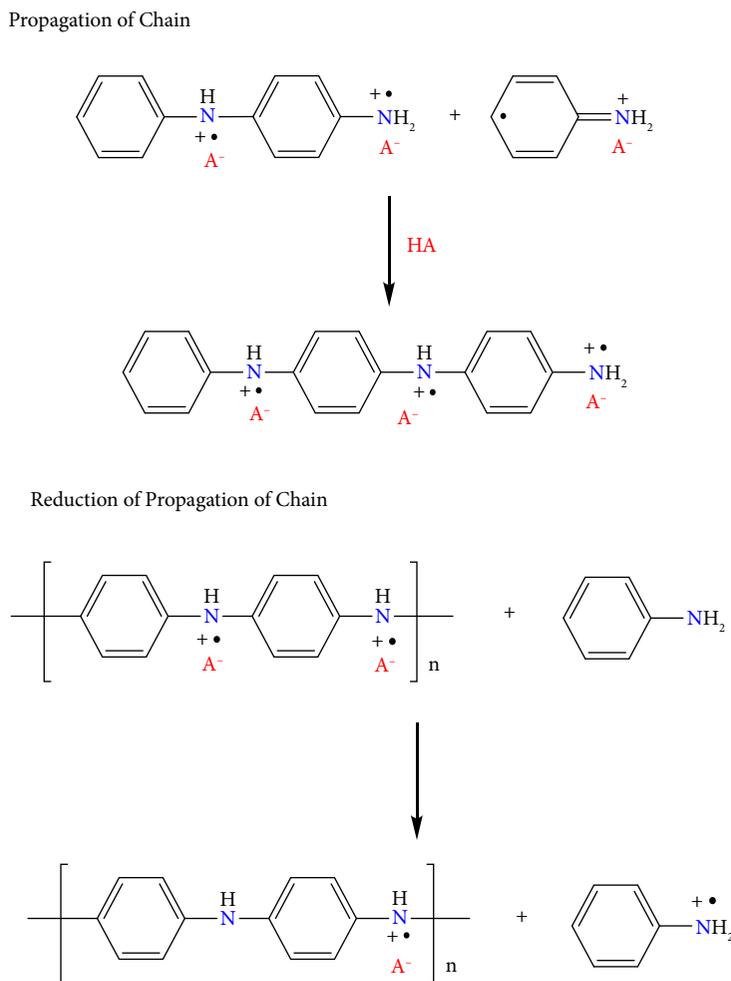


FIGURE 5: Chemical (oxidative) polymerization of aniline [39, 50].

mechanism similar to the mechanism of polymerization of aniline by electrochemical method as described previously [46–48]. The first stage includes the formation of the radical cation of aniline by capturing nitrogen electrons with ammonium persulfate [49].

In the second step, the reaction occurs between the nitrogen radical cation with the radical cation in the para-site of another aniline molecule, and the reaction continues (Figure 5). Mostly, the interaction between the radical cation of nitrogen in the first aniline molecule with the radical cation occurs in the *para* site of the second aniline molecule, but sometimes, the reaction occurs with the radical cation in the *ortho* position with the second aniline molecule, and this leads to a distortion of the resulting PANI chains [39].

3.3. Vapor-Phase Polymerization. One of the important processes of PANI synthesis is VPP. This method was used for the first time to deposit conductive PANI on cotton threads in 1998, by saturating cotton threads with ammonium persulfate $(\text{NH}_4)_2\text{S}_2\text{O}_8$, then passing aniline steam over cotton threads to be coated with conductive PANI [51].

VPP technology is used to manufacture ultra-thin polymer films, such as PANI, polypyrrole, polythiophene, and

others. This process is one of the techniques of self-assembly polymerization that takes place under the vapor phase, which can easily prepare thin films of polymers with very high purity and with superconductivity, because they are free of impurities, as well as the possibility of preparing them at the nanoscale [44, 52, 96, 97].

3.4. Photochemically Initiated Polymerization. Photocatalytic polymerization is a promising and attractive method for producing PANI with unique properties and compositions. In addition, it is a very important method for the production of hybrid materials [53]. In 1998 [54] and 2001 [55], Kobayashi and co-workers demonstrated that it is possible to prepare PANI by photopolymerization using single- or bi-layer films containing $[\text{Ru}(\text{bipy})_3]^{2+}$ as a primer and methylviologen (MV^{2+}) as an oxidizer, then irradiated with visible light to give PANI. $[\text{Ru}(\text{bipy})_3]^{2+}$ is irradiated at the 452 nm region, and this leads to the generation of the triple excited state $^*[\text{Ru}(\text{bipy})_3]^{2+}$. The process of electron transfer between $^*[\text{Ru}(\text{bipy})_3]^{2+}$ and MV^{2+} leads to the generation of the strong oxidizer $[\text{Ru}(\text{bipy})_3]^{3+}$, which is responsible for the process of oxidation of aniline and then polymerization of the latter to PANI. In these studies, the formed PANI

was deposited on monolayer or bilayer films to be used later in molecular electronics.

Later, different techniques were used for the irradiation process to form PANI using an external source, including gamma rays [56], microwaves [57], ultraviolet rays [58], and X-rays [59].

3.5. Enzyme-Catalyzed Polymerization. The researchers in the past years have resorted to using enzymes, such as horseradish peroxidase (HRP), as catalysts for the synthesis of some important polymers, such as PANI and poly pyrrole in the presence of oxidants, such as peroxide [60, 61]. This method is considered environmentally friendly, because the peroxide is converted to water, but several studies have produced branched polymers with low molecular weight. To overcome these problems, Samuelson et al. [62, 63] used various electrolytes, such as poly(styrene sulfonate) (PSS) as templates in the HRP-catalyzed PANI preparation mixture to produce regular polymeric chains. PSS plays three roles in this process:

- (i) PSS acts as a template that aligns the aniline monomers before starting the polymerization process in order to arrange the monomers in the desired head-to-tail coupling position.
- (ii) It provides important doping in the process of activating PANI into the form of electrically conductive emeraldine salt.
- (iii) It imparts the water solubility of the (PANI/PSS) product [64].

In this method, the polymerization process takes place in an acidic medium at (pH = 4), in which water-soluble PANI is obtained [65]. The good electrolytes used are poly(vinylphosphonic acid) and DNA, as well as PSS [66, 67].

4. Properties of PANI

4.1. Physical Properties. PANI has various physical properties, depending on its three forms, where these forms show different colors according to their chemical nature. PANI appears white/clear or colorless if the polymer form is leucoemeraldine, and if the polymer type is pernigraniline, it gives blue to violet color, and if the polymer form is emeraldine, it gives blue color if it is basic and gives green color if it is salt form. PANI also has good electrical conductivity only in the emeraldine form, and the other types do not show significant conductivity [37]. As for solubility, all forms of PANI are insoluble in water, acetone, ethanol, and other solvents, but are soluble in dimethylformamide, dimethyl sulfide, and *N*-methyl-2-pyrrolidone [37, 68, 69].

4.2. Optical Properties. The optical properties of PANI are used to study the oxidation level and protonation process of the polymer. Huang and MacDiarmid observed that when leucoemeraldine was converted from its base form to its salt form, a blue shift occurred from 3.94 to 4.17 eV. This shift occurred due to the excitation of electrons between the locally occupied upper molecular orbital (HOMO) to the

lower unoccupied molecular orbital (LUMO), where the absorption peak was observed at 2.1 eV [70]. Barbero and Kötzt studied the optical properties of PANI films deposited on gold, and two absorption states appeared in the UV-visible region of the oxidized state of the polymer, the first at 3.8 eV (which is due to the $\pi-\pi^*$ transition*) and the second at 2.75 eV (which is related to the n-to- π^* transition), as it has one absorption band at 1.5 eV near infrared and is due to the transition entering the band [71].

For emeraldine conductive PANI in its salt form, there are predictions of molecular orbital calculations, and later (ab initio calculations), where these studies show the presence of a single broad polaron band within the band gap. This band is half full, resulting in a signal in the ESR scan. Calculations of this band structure are consistent with the observed UV-visible and infrared spectra [72, 73]. The emeraldine salt in their compact coil structure shows three bands: the first band at 330 nm, which is assigned to ($\pi-\pi^*$ band transitions), and two visible-region bands at 430 nm, which is attributed to ($\pi \rightarrow$ polaron band), and 800 nm, which related to (polaron $\rightarrow \pi^*$) [74].

4.3. Mechanical Properties. PANI shows a noticeable difference in mechanical properties according to the polymerization method from which it was prepared. Emeraldine salts prepared by electropolymerization method are highly porous and have poor mechanical properties. In addition, PANI films prepared by electropolymerization method have poor mechanical properties, so they are not used frequently. As for PANI prepared from solutions, it is more rigid and is widely used as films and fibers, because it has less porosity and better mechanical properties [39]. The first to study the mechanical properties of PANI are Wei et al., where they noticed that a layer of PANI with a thickness of $0.07 + 0.03$ mm at a temperature of 25°C and under nitrogen atmosphere showed a storage modulus of ~200 MPa [75].

4.4. Electrical Properties. PANI is an attractive conductive polymer that is widely used in the electrical field, because it has good electrical conductivity in its emeraldine salt form. When the acid is added with the basic emeraldine of PANI, it works on protonating the imine group's nitrogen, thus converting it into a salt, thus turning from the non-conductive form to the electrically conductive polymer [37]. Generally, the range of PANI conductivity is from $\sigma \leq 10^{-10}$ S/cm (undoped base emeraldine) to $\sigma \geq 10$ S/cm (doped salt emeraldine) [76, 77]. The process of improving the electrical conductivity or increasing the number of different properties of PANI simultaneously is coupled with the doping process of the polymer. In recent decades, many researchers in the world have tended to dope PANI with various classes of chemical compounds for use in different applications. The purpose of this is to improve the electrical conductivity of the new compounds prepared (polymer compounds) to be used in that application [37, 78]. Table 1 shows the electrical conductivity of the items prepared with PANI.

4.5. Anti-Corrosion Properties. Corrosion is a chemical change to the metal due to the environmental influences

TABLE 1: Electrical conductivity of polyaniline and/or its composites.

No.	PANI system	Dopant	Conductivity S/cm	References
1.	PANI-SAP	Sulfonic aminophenol	0.13×10^{-3}	[79]
2.	PANI-H ₂ SO ₄	Sulfuric acid	1.09	[80]
	PANI-NA	Nicotinic acid	0.63	
3.	PANI-MNA	2-Methyl nicotinic acid	0.089	[81]
	PANI	Molybdenum disulfide	1.09×10^{-1}	
	MoS ₂ /PANI-8		2.5×10^{-1}	
4.	MoS ₂ /PANI-38		2.38	[82]
	FTO/PANI	Fluorine doped tin oxide (FTO) and graphene (GR)	0.09	
	FTO/PANI-GR 1 mg		0454	
5.	FTO/PANI-GR 3 mg		4.76	[83]
	GO-PANI	Graphene oxide	0.1	
6.	PANI	Carbon black (CB)	0.01	[84]
	PANI/CB	Graphite (GR)	0.03	
	PANI/GR	Multi-wall carbon nanotubes (MWCNTs)	0.07	
7.	PANI/MWCNTs		2.28	[85]
	PANI		0.50	
8.	PANI/MWCNTs	MWCNTs	1.95	[86]
	PANI-Au	Gold (Au)	2.13×10^{-4}	
9.	PANI-Au-CSA	Camphor sulfonic acid (CSA)	5.42×10^{-3}	[87]
	PANI	Tin dioxide (SnO ₂)	0.21	
10.	PANI-SnO ₂ (0.02 g)		2.6×10^2	[88]
	PANI-SnO ₂ (0.05 g)		3.1×10^2	
11.	PANI	Silver NPs (Ag)	3.3×10^{-3}	[89]
	PANI/Ag		5.2×10^{-2}	
12.	PANI 9%-DBSA/DVB	Dodecylbenzenesulfonic acid (DBSA)	0.01	[90]
		Divinylbenzene (DVB)		
13.	PANI 13%-DBSA/PC	Polycarbonate (PC)	0.01	[91]
14.	PS/(PANI-DBSA) 30%	Polystyrene (PS)	0.001	[92]
	PANI 0%/PVA-g-PAA	Poly(vinyl alcohol) (PVA)-g-poly(acrylic acid) (PAA)	0.01	
15.	PANI 5%/PVA-g-PAA		0.04	[93]
16.	RGO/PANI/EVA	Reduced graphene oxide (RGO) and ethylene vinyl acetate (EVA)	1.7×10^{-1}	[94]
17.	PANI-TiO ₂	Titanium dioxide (TiO ₂)	1×10^{-5}	[95]
	PANI		2.6×10^{-2}	
18.	PANI-TiO ₂ (15%)	Titanium dioxide (TiO ₂)	3.8×10^{-2}	[96]
	PANI-TiO ₂		4.8×10^{-2}	
19.	PANI-NC	Nanoclay (NC)	2.143	[97]
19.	PVA/PANI/Ni	Poly(vinylalcohol) (PVA) and nickel nanoparticles (Ni)	4.9	[97]

surrounding it, and it is one of the major problems facing the various processes that use the metal as a base element in their applications. Many materials have been used to end or reduce the corrosion process, and one of these materials is PANI, which was first reported in the 1980s [41]. PANI was used to effectively protect steel from oxidation (corrosion) when it was used with fuel cells at the same time. Research and studies have proven that preparing layers of sulfate with PANI is much better and more suitable for reducing the corrosion of steel than layers of PANI with phosphate [98]. The materials

prepared from PANI molybdate and PANI tungstate as coating materials to reduce steel corrosion were studied by Kamaraj et al. [99]. Likewise, with nickel-plated steel, the PANI coating showed great protection for this steel from corrosion [100]. In addition, Mirmohseni and Oladegaragoze used PANI-coated iron in different corrosive environments to study the corrosion potential and corrosion current [101].

4.6. Semiconducting Properties. PANI is considered one of the most promising materials in the field of electronics due

TABLE 2: Summary of some polyaniline nanocomposites synthesis with metal nanoparticles.

No.	PANI nanocomposite type	Dopant	Preparation method	References
1.	PANI/Au	Au	In situ interfacial polymerization	[123]
2.	PANI/Ag	Ag	Oxidative polymerization	[124]
3.	PANI/Ag	Ag	In situ photo-redox mechanism	[125]
4.	PANI/Ag	Ag	In situ polymerization	[126]
5.	PANI/Fe	Fe	Sonochemical method	[127]
6.	PANI/Ni	Ni	Oxidative polymerization	[128]
7.	PANI/Cu	Cu	Electrochemical polymerization	[129]
8.	PANI/Pd	Pd	Templating method	[130]
9.	PANI/Pt	Pt	In situ interfacial polymerization	[131]
10.	PANI/Pr	Pr	Oxidative polymerization	[132]
11.	PANI/Se	Se	Oxidative polymerization	[133]
12.	La/Cd/PANI	La/Cd	Graft copolymerization method	[134]
13.	PANI/CdS	CdS	Facile chemical methods	[135]
14.	PANI/PbS	PbS	Chemical oxidation technique	[136]
15.	PANI/Au Pd	Au Pd	Oxidative polymerization	[137]

to its good conductivity of electrons. The results of electrical and thermal conductivity of the ionic liquid of PANI showed that this polymer is a p-type semiconductor, and the diode of PANI was prepared by Yakuphanoglu and Şenkal [102]. On the other hand, PANI films doped with boric acid were prepared, then the electrical properties of these films were examined. It was found that PANI films doped with boric acid show semi-conductive properties, and its electrical conductivity was found 1.02×10^{-4} S/cm at room temperature. The optical band gap of the PANI films doped with boric acid was 3.71 eV. Because of these semiconducting properties, PANI is widely used in organic field transistors and in solar batteries [103].

4.7. Capacitive Properties. PANI has been widely used in recent years in various electrical and electronic devices, especially electrical storage devices, such as the super capacitor. PANI has contributed to improving the properties of these energy-saving devices due to the rapid oxidation and reduction reactions that occur in a large part of the polymer material. PANI gives a good capacitive response and produces superior specific energies compared to double-layer capacitors. Moreover, PANI has a stronger effect than inorganic batteries and also has remarkable electrical conductivity. However, capacitors made of pure PANI experience contractions in the charge-discharge cycle [41, 104]. To overcome these problems and enhance the properties of the capacitors, researchers resorted to using other compounds with PANI, such as PANI-graphene [105–107], PANI-carbon nanotubes [108–110], PANI-manganese oxide [111], PANI/MoS₂-MnO₂ [112], PANI-silver-manganese dioxide [113], and others. These modifications of the electrodes containing PANI proved to be of great improvement in the increase of energy density, stability, and strength [41].

4.8. Magnetic Properties. Due to its high spin density, PANI shows good interest in magnetic behavior and has been used

in many studies [114]. The paramagnetic nature is closely and directly related to the magnetic and electrical properties of electrically conducting PANI, which is caused by the presence of hydrochloric acid as dopant with PANI as a result of the oxidation process [115].

The polymeric composite nanotubes (PANI-Fe₃O₄) have been prepared in [116], and this composite exhibited good magnetic behavior and significant magnetic properties. Similarly, the self-assembly method was used to prepare the polymeric composite (PANI-Fe₃O₄) nanorods. In addition, because hydrogen bonding in methanol and PANI structures actually affects the polarization, it has been observed that there are good magnetic properties of iron in PANI [41].

The rotation sensitivity of PANI refers to the magnetic centers that are found in charge carriers that are implanted in metal fields as well as those along polymer chains. In this spin, the electron comes into contact with the samples, and this leads to clearly showing anti-ferromagnetic properties, such as paramagnetic sensitivity [41, 117].

5. Nanocomposites of PANI

A composite is a new substance resulting from the mixing of two or more materials that are significantly different from each other in properties. The resulting new materials (composites) have different properties or improved properties from those of the parent materials. The idea is to form a new material (composite) to bring out new, diverse, and remarkably progressive properties [118]–[120]. Although PANI exhibits attractive qualities and outstanding properties, there are some limitations in some applications [121]. Making some improvements on PANI, such as combining it with other materials to form polymeric composites, is very important to give higher efficiency in different applications. The process of mixing materials with polymers is the result of polymeric composites, so if the added material is in the

TABLE 3: Summary of some polyaniline nanocomposites synthesis with metal oxide nanostructures.

No.	PANI nanocomposite type	Dopant	Preparation method	References
1.	PANI/TiO ₂	TiO ₂	In situ deposition oxidative polymerization	[138]
2.	PANI/MnO ₂	MnO ₂	Simultaneous-oxidation route	[7]
3.	PANI/MgO	MgO	In situ polymerization method	[139]
4.	PANI/ZnO	ZnO	In situ chemical oxidation method	[140]
5.	PANI/NiO	NiO	Electrodeposition method	[141]
6.	PANI/Fe ₂ O ₃	Fe ₂ O ₃	In situ polymerization	[142]
7.	PANI/ γ -Fe ₂ O ₃	γ -Fe ₂ O ₃	In situ polymerization	[143]
8.	PANI/ α -Fe ₂ O ₃	α -Fe ₂ O ₃	In situ polymerization	[144]
9.	PANI/CuO	CuO	In situ polymerization	[145]
10.	PANI/Co ₃ O ₄	Co ₃ O ₄	In situ polymerized method	[146]
11.	PANI/V ₂ O ₅	V ₂ O ₅	Chemical oxidation method	[147]
12.	PANI/RuO ₂	RuO ₂	Chemical bath deposition (CBD) method	[148]
13.	PANI/SnO ₂	SnO ₂	Chemical oxidative polymerization	[149]
14.	PANI/Sm ₂ O ₃	Sm ₂ O ₃	In situ oxidative polymerization	[150]
15.	CeO ₂ /PANI	CeO ₂	Chemical oxidative polymerization	[151]
16.	PANI/Al ₂ O ₃	Al ₂ O ₃	Surface initialized polymerization (SIP) method	[152]

TABLE 4: Summary of some polyaniline nanocomposites synthesis with carbon nanocompounds.

No.	PANI nanocomposite type	Dopant	Preparation method	References
1.	PANI/SWCNT	SWCNT	Oxidative polymerization method	[153]
2.	PANI/f-SWCNT	f-SWCNT	Electrochemical polymerization	[154]
3.	PANI/MWCNT	MWCNT	Oxidative polymerization method	[155]
4.	PANI/MWCNT	MWCNT	In situ enzymatic aniline polymerization	[156]
5.	MnO ₂ /PANT/MWCNT	MnO ₂ /MWCNT	In situ chemical oxidation polymerization	[157]
6.	MWCNTs/PANI/TiO ₂	MWCNT/TiO ₂	One-pot polymerization process	[158]
7.	CuO@NiO/PANI/MWCNT	CuO@NiO/MWCNT	Oxidative polymerization method	[159]
8.	PANI/GO	GO	In situ polymerization technique	[160]
9.	CuNPs-PANI-GO	CuNPs-GO	In situ oxidative polymerization	[161]
10.	Ag ₃ PO ₄ -PANI-GO	Ag ₃ PO ₄ -GO	Interfacial polymerization	[162]
11.	PANI-RGO-ZnO	RGO-ZnO	In situ oxidative polymerization	[163]
12.	PANI/graphene	G	In situ polymerization	[164]
13.	Pt-PANI-graphene	Pt-G	In situ oxidative polymerization	[165]
14.	Polyaniline/fullerene C ₆₀	C ₆₀	Copolymerization of aniline	[166]
15.	Polyaniline/graphite oxide	Graphite oxide	Interfacial polymerization	[167]

nanoscale dimensions, the new material is called the polymeric nanocomposite. It has been observed that many materials were mixed with PANI to form PANI composites. The additives added to PANI to form PANI composites may be in the form of metals, metal oxides, carbon nanostructures, polymers, or various organic compounds [122]. Table 2 shows the preparation of PANI nanocomposites with metal nanoparticles.

PANI composites were also synthesized with metallic nano-oxides due to their great importance in many applications, such as electrical storage devices, water purification, electronic devices, supercapacitors, sensors, solar cells, and others. Table 3 shows the preparation of PANI nanocomposites with metal oxides and the method of their preparation.

On the other hand, PANI was extensively synthesized with carbon nanocompounds, due to the high properties demonstrated by these prepared nanocomposites compounds. The prepared PANI nanocomposites were used in many different applications and showed high efficiency in the applications used in them. Table 4 shows some PANI nanocomposites with different carbon nanocompounds.

In addition, there is a huge amount of polymeric nanocomposites of PANI in different shapes and structures and with different applications and polymerization techniques, and this is clear evidence of the importance of PANI and its wide uses in the experimental and the industrial field.

TABLE 5: Some of the different studies used in the applications of supercapacitor based on polyaniline or its nanocomposites.

PANI nanocomposites	Electrolyte	Specific capacitance	Cycle stability	Energy and power density	References
PANI	1 M H ₂ SO ₄	503 F/g	85% (10,000)	96 W h/kg and 8.88 kW/kg	[172]
PANI nanowires	1 M H ₂ SO ₄	742 F/g	92% (1500)	68 W h/kg and 16 kW/kg	[173]
PANI nanofibers	1 M H ₂ SO ₄	548 F/g	75% (1000)	36 W h/kg and 0.12 kW/kg	[174]
Sulfonated PANI	1 M H ₂ SO ₄	1107 F/g	—	31.4 W h/kg and 14.8 kW/kg	[175]
PANI/G/CNT	1 M H ₂ SO ₄	510 F/g	91% (5000)	20.5 W h/kg and 25 kW/kg	[176]
GO/PANI	PVA/H ₃ PO ₄	1095 F/g	—	24.3 W h/kg and 28.1 kW/kg	[177]
PANI/G	EMITFSI/PVDF-HFP	87.8 mF/cm ⁻² at 0.22 mA/cm ⁻²	100% (10000)	12.2 mW h/cm ⁻² and 226.4 mW/cm ⁻²	[178]
Carbonized iron/PANI/G	1 M NaNO ₃	69.9 F/g	91% (10000)	68 W h/kg and 718.2 W/kg	[179]
G/PANI/MnOx	1 M Na ₂ SO ₄	955 F/g	89% (1000)	61 W h/kg and 4.5 kW/kg	[180]
Polyaniline/G/Fe ₂ O ₃ hydrogel	1 M H ₂ SO ₄	1124 F/g	82% (10000)	14.4 W h/kg and 58 W/kg	[181]
PANI/TiO ₂ /GO	1 M H ₂ SO ₄	430 F/g	—	—	[182]
PANI/SWCNT	1 M H ₂ SO ₄	485 F/g	—	228 W h/kg and 2250 W/kg	[153]
PANI/MWCNT	1 M NaNO ₃	328 F/g	94% (1000)	—	[183]
PANI/MnO ₂	0.1 M Na ₂ SO ₄	715 F/g	96.5% (5000)	200 W h/kg	[184]
MnO ₂ /PANI/Hollow mesoporous c	PVA-KOH	248.5 F/g	97.7% (5000)	88.4 W h/kg and 800 W/kg	[185]
PANI/SnO ₂	1 M H ₂ SO ₄	305 F/g	95.5% (500)	42 W h/kg	[186]
PANI/V ₂ O ₅	5 M LiCl	443 F/g	92% (5000)	69 W h/kg and 0.72 W/kg	[187]
PANI/MoS ₂	1 M H ₂ SO ₄	575 F/g	98% (500)	265 W h/kg and 18 W/kg	[188]
MnO ₂ /PANI/MWCNTs	1 M KOH	395 F/g	72% (1000)	—	[157]
PANI coated RuO ₂ /TiO ₂	0.1 M H ₂ SO ₄	67.4 F/g	81.6% (10,000)	3.37 W h/kg and 60 W/kg	[189]
PANI-RGO-ZnO	1 M H ₂ SO ₄	40 F/g	86% (5000)	5.61 W h/kg and 403 W/kg	[163]
CuO@NiO/PANI/MWCNT	3 M NaOH	1372 F/g	83% (1500)	—	[159]
PANI/rGO-Co ₃ S ₄	1 M H ₂ SO ₄	767 F/g	81.7% (5000)	61.2 W h/kg and 8.8 kW/kg	[190]
PANI/MoO ₃ /G	1 M H ₂ SO ₄	593 F/g	92.4% (1000)	55.5 W h/kg and 1678.99 W/kg	[191]

6. Application of PANI

Conductive polymers, since their discovery until now, have shown very unique properties due to their distinctive electronic arrangement, chemical nature, and due to their diverse physical specifications, and this makes them essential materials in many different applications. For example, PANI is a good electrical conductive polymer, and due to its low cost, ease of preparation, unique properties, and non-polluting of the environment, it attracts the interest of many researchers for its use in many applications. The most important applications in which PANI is used as a main material or as a secondary material are as follows.

6.1. In Supercapacitor. The world economy has played a very important role in the use of multiple sources of energy, such

as natural gas, fossil fuels, and gas. The use of this fuel provides good energy, but it causes many environmental problems. Therefore, it has become very necessary to replace the traditional energy system with alternative energy sources. For this, scientists have always sought to develop and produce environmentally friendly, high-energy, and renewable energy sources, such as water energy, supercapacitors, wind energy, and fuel cells. Super capacitors are considered one of the most promising energies and of great commercial importance in the future markets, because they are used in many different applications, such as electrical and electronic devices and wearable devices. The main difference between super capacitors and conventional is that super capacitors have a capacity for storing energy 1000 times more than dielectric capacitors, as well as their long life, and show high-energy density and have fast charging and discharging [168].

TABLE 6: Some different studies on sensors based on polyaniline or one of its nanocomposites.

Sensory system	Analytes	Sensitivity % at ppm	Range (ppm)	Response time	Recovery time	Detection limit	References
PANI	NH ₃	140% for 100 ppm	1–2000 ppm	60 s	240 s	—	[201, 203]
PANI-MWCNTs	NH ₃	320% for 100 ppm	0.9	20 min	40 min	50 ppm	[204]
PANI-MWCNT	CO	6.8–26.7%	—	76 s	210 s	—	[205]
PANI-SWCNT	NH ₃	5.8%	0.1–100	—	—	0.05 ppm	[206]
PANI-SWCNT	NO ₂	1.9%	0.5–80	—	—	0.5 ppm	[206]
PANI-SWCNT	H ₂ S	3.6%	2–100	—	—	0.5 ppm	[206]
PANI/Co ₃ O ₄	CO	8.6%	—	140 s	90 s	—	[207]
PANI/TiO ₂ nanorods	NH ₃	75% for 100 ppm	1	40 s	400 s	20 ppm	[208]
PANI-SnO ₂ NPs	NH ₃	8% for 100 ppm	0.08	10 s	150 s	100 ppm	[209]
SnO ₂ /rGO/PANI	H ₂ S	9.1% for 0.1 ppm	0.1	—	—	0.05 ppm	[210]
PANI/TiO ₂ nanofibers	NH ₃	180% for 0.2 ppm	0.33	20 s	50 s	25 ppb	[211]
PANI/TiO ₂	Acetone	0.5%	15–50	—	—	10 ppm	[212]
PANI/TiO ₂	Ethyl acetate	0.06%	10–80	—	—	10 ppm	[212]
PANI/TiO ₂	Toluene	1.4%	2–35	—	—	2 ppm	[212]
PANI/TiO ₂	Ethanol	2.1%	1–35	—	—	0.5 ppm	[212]
PANI/AuNPs	H ₂ S	—	0.1–8000	—	—	0.1 ppm	[194]
PANI/rGO	NH ₃	50%	0.01	1000 s	50 s	5 ppm	[213]
PANI/Ag NPs	NH ₃	200%	1.2	20 min	30 min	100 ppm	[214]
PANI/Au/Al ₂ O ₃	Acetone	0.06	30–300	—	—	30 ppm	[215]
PANI-PMMA fibers	Ammonia	550% for 30 ppm	27	200 s	300 s	1 ppm	[216]
PANI/In ₂ O ₃	NH ₃	90% for 93 ppm	0.7	10 s	60 s	23 ppm	[217]
PANI/acrylic acid	Ammonia	2%	1–600	—	—	1 ppm	[218]

Supercapacitors can be divided into three main types, which are as follows: (i) double-layer electrostatic capacitors, whose electrodes are mostly carbon materials, such as carbon, carbon nanotubes, and graphene, (ii) pseudo-electrochemical capacitors, whose electrodes are mostly made of conductive polymers and metal oxides, and (iii) hybrid pseudocapacitors, which are a mixture between the above two types [121, 169].

The electrode materials used in supercapacitors have a very important role on the efficiency of the storage device and the energy conversion process. The electrodes used in supercapacitor devices depend on the use of carbon materials, metals, and conductive polymers [170]. Although supercapacitors using various carbon materials exhibit significant energy density, good conductivity, and adequate durability, their energy density is low. As for metals, they show very appropriate electrochemical properties when used in super-capacities, but their abundance in nature, difficulty in obtaining them, and high cost limit their use for this purpose. Because of its different oxidation states, high conductivity, and specific capacitance, PANI can be used with great success in supercapacitors. In addition, the flexible electrochemical properties of PANI make it very suitable to be strongly used in the manufacture of supercapacitor electrodes. The electrochemical properties of supercapacitors in which PANI is used as a base material in the electrodes depend on the properties of the PANI itself, such as synthesis method, chemical/physical properties, dopant, and nanostructure of PANI [171]. Various carbon compounds were used with PANI to prepare the supercapacitors, such as gra-

phene, fullerene, carbon nanotubes, and graphene oxide. Metal nanomaterials with PANI were also used for the same purpose. Table 5 shows the use of various types of materials with PANI to prepare the super capacitors.

There are also a very large number of polymeric nanocomposites of PANI used in supercapacitors, and they gave good efficiency while using as supercapacitors.

6.2. In Sensors. The device that can show the occurrence of various changes in its environment and detect the presence of other things is called a sensor. The common sensors at present are composed of organic or inorganic semiconducting materials or films [192]. One of the materials that have gained great demand in recent times for the manufacture of sensors is conductive polymers. PANI is one of these polymers, which has received great interest as sensors due to its different structures and morphologies in which it can be prepared, such as microstructures, nanowires, nanotubes, nanoparticles, and nanosheets [193]. Using PANI or one of its nanocomposites, various sensors have been manufactured, such as chemical and biological sensors. Researchers have tried to prepare PANI with different nanostructures, such as nanofibers, nanowires, and nanotubes; then, these nanostructures were used in gas sensors because of their very high surface area and the possibility of gas emission. Pure PANI cannot detect H₂S gas alone, because H₂S gas molecules cannot change the conductivity of PANI, so gold nanoparticles are grafted with PANI to form gold nanocomposites with PANI, and this new system can easily detect H₂S gas, and the

response rate reaches very low concentrations (0.1 ppb) [194]. PANI nanostructures were also used to sense the presence of ammonia, hydrochloric acid, hydrazine, chloroform, carbon oxides, and methanol Table 6, and this led to a noticeable structural change in the PANI nanostructure chains [38]. The sensing property of PANI can be improved by adding different nanomaterials to form PANI nanocomposites, as this reduces the sensing time and leads to the detection of materials or gases even in very low concentrations [193, 195–202].

7. Conclusions

Providing polymeric composites with metals or their oxides in addition to various other composites in order to improve some properties is the subject of all investigations now. Because PANI originally has unique electrical properties, it always attracts the attention of researchers in their studies as well as in various applications, the most important of which is electrical applications. We concluded during this study that it is possible to improve the electrical properties of PANI to be used in sensor applications as well as in other applications.

The recent discoveries in the chemistry of PANI made researchers to devote their efforts to research in this field. It appeared to PANI with important specifications that made it preferred over the rest of the other polymers. One of the most important properties of PANI is its electrical conductivity and its various applications, which opened new horizons for researchers to research in this field. It was found that PANI possesses real electrical properties that made it a cornerstone in many applications that could be of great help to humanity in their lives, including super capacitors, solar cells, all kinds of sensors, batteries of all kinds, and fuel cells. It was also found that the electrical properties of PANI are significantly improved when it is grafted with some nanomaterials to form PANI nanocomposites. We concluded, during this study, that it is possible to improve the properties of PANI by grafting it with other materials, especially nanomaterials, to form polymeric nanocomposites of PANI, and that the latter can improve the applied properties of supercapacitors, gas sensors, and others, as documented in this study.

Data Availability

The data used to support the findings of this study are included within the article.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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