Conducting polymers for flexible devices

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ABSTRACT

Electronic wearable devices of the next generation, which are mechanically flexible and stretchable and need to fit the human body conformally, are in high demand. Energy harvesting systems have witnessed a huge increase in the use of organic and polymeric semiconducting materials due to their cheap cost, large-area manufacturing, high flexibility, low toxicity, and tunable electronic properties. The extraordinary characteristics of conductive polymers, such as lightweight, flexibility, and non-toxicity, make them intriguing options for next-generation stretchy, flexible, and wearable electronic systems.

INTRODUCTION

Flexible electronic materials and technologies have received much attention due to their numerous applications in various fields, including flexible displays, soft robotics, healthcare monitoring, human-machine interfaces, and other areas.[1-3] For electrical devices to be flexible, the electrodes and active components have to be mechanically stretchable. The Young modulus of metals is significantly higher than that of elastomers, which are often utilized as the substrate for flexible electrical devices even though metals may be relatively thin and still have significant mechanical flexibility. The low transparency of bulk metals in the visible range of light makes them unsuitable for use as transparent electrodes in optoelectronic devices like solar cells and light-emitting diodes (LEDs). Flexible electronic devices can use solar cells and flexible LEDs for their displays and power sources. An optoelectronic device must have a transparent electrode on at least one electrode to produce or capture light. However, oxides like indium tin oxide (ITO) are most often used as transparent conductors. To serve as the flexible transparent electrode, they are too fragile. Inherently conductive polymers (CPs) and conductive nanomaterials are examples of potential flexible transparent conductors.[4-6] Significant research has been done on the flexible transparent electrode for optoelectronic devices using metal nanowires, particularly silver (Ag) nanowires and carbon nanotubes (CNTs). In addition to their inherent conductivity, their resistance is also influenced by the contact resistance between the nanowires or nanotubes. However, the sheet resistance of CNT film is too low, which makes it difficult for practical use, whereas Ag nanowire electrodes have problems such as adhesion to the substrate, excessive roughness, and performance deterioration brought on by surface oxidation.[7, 8] A fascinating alternative for the transparent electrode is graphene, which has a high degree of mechanical flexibility and extreme transparency.[9] However, a thin graphene layer with remarkable homogeneity across a vast area is challenging to manufacture. Given its potential for high mechanical flexibility and intrinsic conductivity, conducting polymers are exciting candidates for the electrode.

The delocalization of electrons allows some conjugated polymers, referred to as conducting polymers, to conduct electricity. This charge mobility leads to exceptional electronic properties including wide surface area, electrical conductivity, strong electron affinity, low energy optical transition, and mechanical flexibility. In 1977, The electrical conductivity of these polymers was initially noted in polyacetylene in 1977 by Shirakawa et al.[10] Following that, these polymers drew much interest from academics and scientists because of their significant economic value, excellent environmental stability, tailored optoelectronic capabilities, practical mechanical qualities, and a host of other factors. They include polyaniline (PANI), polypyrrole (PPy), polythiophene (PTh), poly(3,4-ethylenedioxythiophene):polystyrenesulfonate(PEDOT:PSS), poly(naphthylamine)(PNA), poly(o-phenylenediamine) (PoPDA) and polycarbazole (PCz), polyanisidine (PANIS).[11-13] These have numerous uses, such as biomedical materials, optoelectronic devices, sensing, electrochromic devices batteries, environmental cleanup and imaging devices [14]. Alternating single and double (or triple) bonds in carbon-based compounds, which are electrically conductive polymers, allow for charge transfer.

The doping level, macromolecular architecture, and crystallinity of these materials are important aspects of their electronic state density. Extending the degree of polymerization π of conjugated polymer chains results in a broadening of the orbitals with considerable overlapping, which causes a shift in state density from isolated states to a band-like structure. As a result, π conjugated polymers can be used to create electronic devices ranging from semiconductors, semimetals to metals. Heeger Alan J discovered the inorganic material $(SN)_x$ sulfur nitride metal, which becomes more electrically conductive when bromine is doped.[15] This discovery encouraged study on polyacetylene and resulted in the discovery of bromine-doped polyacetylene, whose conductivity is a million times greater than that of pure polyacetylene.[15] Traditional polymers are constructed from tens of thousands to millions of monomer units. A conjugated polymer chain is nonetheless stiff and soluble in solvents despite having fewer monomer units than conventional polymer chains. It possesses mechanical characteristics because the single and double bonds alternate. The linked side chains of conducting polymers, which also give them their mechanical, electrical, and optical characteristics, are primarily responsible for their solubility and processability.[16] Conducting polymers are partially crystalline and amorphous. Both localised and delocalized forms of conducting polymers are possible, and the disorder significantly affects the delocalization of π-bonds. This delocalization is required for the generation of charge carriers like polarons, bipolarons, solitons, and other particles involved in the conversion of insulators into metals. In their pure state, conjugated polymers act as an insulator to semiconductors, and their conductivity improves with dopant concentration.[17] In conjugated polymers, the -bond self-localizes to undergo nonlinear excitation as polarons, solitons, bipolarons, etc. when doped or exposed to light. A metallic state is reached after the polymer undergoes a transformation from a nonlinear excitation state.[18]

TYPES OF CONDUCTING POLYMERS

A. Polyacetylene π

The Polyacetylene (PA) conventional conjugated polymer, a linear polyene chain [-(HC=CH)-], has intriguing features as shown in figure (1), make it an excellent candidate for application in electrical devices. Producing supramolecular assemblages, helical graphitic nanofibers, chiral recognition, gas permeability and liquid crystallisation are a few of these.[19, 20] Other characteristics include high electrical conductivity and photoconductivity. PA is one of the most elementary organic polymers that can exhibit conductivity similar to metal.[10] Since the chemical structure of PAs contains repeating units of two hydrogen atoms, substituting other molecules for hydrogen atoms to produce mono- or di-substituted PAs makes it simple to add pendant groups to the chain.[19]

According to their conformation, cis- and trans-PA's non-doped electrical conductivity is 10^{-9} S/cm and 10^{-6} S/cm, respectively.[21] By comparison, PAs, through p- or n-doping, can reach an almost metallic conductive level (10^{4}-10^{5} S/cm).[22, 23] Although PA has a promising potential to conduct electricity, its processing challenges and significant instability, even at ambient temperature have severely restricted its use in real-world applications.[24] In comparison to other CPs with lower electrical conductivity but greater stability and processability, PA has been employed less in the design and development of flexible electronics. PA is synthesized via a variety of techniques, including precursor-assisted synthesis, catalytic and non-catalytic polymerization, and others.[18] Acetylene and other monomers are often polymerized using catalytic polymerization techniques like Ziegler-Natta or Luttinger catalysis to synthesize PA and oligomers like cyclooctatetraene and vinyl acetylene.[25]. High solubility in organic solvents and excellent selectivity make Zeigler-Natta catalysts, which are made of $Ti(0\text{-}n\text{-}C_4H_9)$ and $(C_2H_5)_3$ Al, the perfect catalysts to generate extremely crystalline PA films.[25]

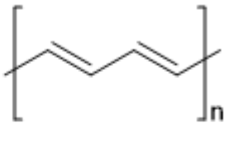

FIGURE 1: STRUCTURAL ILLUSTRATION OF POLYACETYLENE

B. Polyaniline

One of the most promising conjugated CPs is polyaniline (PANI), shown in Figure 2, because of its excellent environmental stability, high processability, high and tunable electrical conductivity and optical characteristics.[26] Dopant concentration and pH have a major impact on PANI conductivity; at pH < 3, it can exhibit conductivity akin to metal.[27] The three distinct forms of PANI are leucoemeraldine $[(C_6H_4NH)]_n$, emeraldine $[([C_6H_4NH]_2[C_6H_4N])_2]_n$, and pernigraniline $[(C_6H_4N)]_n$ based on the idealised oxidation states of the aniline monomers.[28] PANI is oxidised in the pernigraniline base (blue/violet). The leucoemeraldine base (white/clear) is completely reduced, whereas the emeraldine (salt-green/base-blue) is partially oxidised. Emeralidine form PANI is more conductive and stable at room temperature. The leucoemeraldine and pernigraniline forms exhibit poor conductivity even with doping. Immersing the emeraldine base in an aqueous acidic solution will change the PANI conductivity, which is highly dependent on the manufacturing method.[29] Hydrogen bonding interactions between neighboring chains and the rigid polymer backbone of emeraldine base state makes it weakly soluble and challenging to process. The instability of it at melt-processing temperatures further limits its range of feasible uses. Functionalized PANI and other PANI derivatives are frequently used for developing flexible electronics.[30] PANI has received significant attention in the design and development of commercial technologies, exceptionally flexible electronic devices, in a variety of fields, including organic electronics[31], biosensors, chemical sensors, photovoltaic cells, electro-rheological material corrosion devices, solar cells and organic light-emitting diodes.[32, 33] This is a result of its low cost, great optical and electrical qualities, excellent environmental stability and strong mechanical and anti-corrosion capabilities.

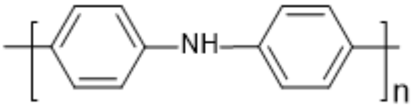

FIGURE 2: STRUCTURAL ILLUSTRATION OF POLYANILINE

C. Poly [3,4-(ethylenedioxy)thiophene]

Poly [3,4-(ethylenedioxy)thiophene] (PEDOT) in figure (3) is a common CP due to its ease of production, high conductivity, superior air stability and optical clarity.[34] In PEDOT crystals, inclined stacks with electrons that are lighter than holes are arranged in monoclinic lamellar structures.[35] According to the PEDOT structural model, the pseudo-orthorhombic unit cell comprises four monomers and one tosylate ion, and its lattice parameters are, 14.0, 6.8 and 7.8Å respectively.[36] The advantages of PEDOT have led to its widespread application in the manufacturing of a variety of flexible devices for sensors, energy conversion, and bioelectronics.[37] PEDOT's hydrophobic nature involves the use of hydrophilic surfactant additives, such as poly(styrenesulfonate) (PSS), to increase its aqueous processability as a thin film. Because of the interaction of the long, water-soluble PSS molecular chains with the short, insoluble PEDOT chains through columbic interactions that result in the formation of grains, PEDOT has efficient water dispersion. PEDOT: PSS grains (30–50 nm in size) are built of tangles made up of several PEDOT segments and a single PSS chain. A number of processing methods and doping chemicals have been used to minimize excess PSS, induce phase separation or morphological rearrangement, and increase the electrical conductivity of PEDOT: PSS.[38] In order to improve the conductivity of PEDOT:PSS films, polar solvents such as dimethyl sulfoxide (DMSO), ethylene glycol and co-solvents or acids like sulfonic acid, chloroplatinic acid, and mineral acids were utilized.

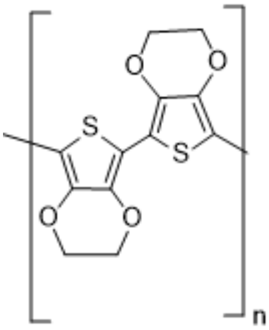

FIGURE 3: STRUCTURAL ILLUSTRATION OF POLY [3,4-(ETHYLENEDIOXY)THIOPHENE]

D. Polypyrrole

A heterocyclic positively charged CP containing N atoms, polypyrrole (PPy) in figure (4), can become completely insulating when sufficiently oxidized.[39] Aqueous solutions and organic electrolytes both increase PPy's electroactivity. PPy is one of the most exciting CPs because of its monomer (pyrrole), which is readily oxidized, widely available, light weight, water-soluble, inexpensive, and biocompatible. This makes it even more fascinating..[40] High conductivity, strong redox characteristics and good environmental stability are also present. This has led to PPy being studied for a variety of applications, including biochemical and electrochemical energy devices. In addition, PPy is more flexible than the bulk of other CPs and may be combined with other nanostructured materials, including graphene or other carbon compounds, to produce nanocomposites. These materials can have much improved properties because of their increased ion diffusion rates and larger contact surface areas. PPy is a major competitor to address the need for flexible and portable electronic devices because of these exceptional qualities.[41]

The alternating single and double bond structure of PPy causes a minor delocalization of the electron density within the molecule, which may be advantageous.[42] A significant band gap energy of 3.16 eV exists in pristine PPy, an insulator.[43] Doping techniques using chemicals and electrochemistry have been used to increase PPy conductivity. The neutral polymer chain's structure can be changed from benzenoid (aromatic) to quinoid in these doping techniques by removing an electron by PPy oxidation. Doping can transform PPy into an ionic complex with additional counter ions and cations. The most popular PPy synthesis techniques employ chemical and electrochemical polymerization. Other cutting-edge polymerization methods, like electrospinning, micro-emulsion, ultrasonic irradiation, mechano-chemical polymerization, vapor-phase polymerization and photopolymerization, are now attracting people's attention to produce highly conductive PPy.[44]

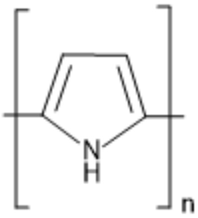

FIGURE 4: STRUCTURAL ILLUSTRATION OF POLYPYRROLE

E. Polythiophene

The polythiophene (PT) in Figure (5) is an important CP due to its simple structure, good stability and a number of optoelectronic properties. Due to structural change and solution processability, PT and its variants in both the undoped and doped states have generated significant attention in sensing device applications.[45] Additionally, it displays a strong affinity for adsorption on the electrode surface as well as a selective barrier effect against certain chemicals. Dopants or side-chain groups with a band gap between 3 and 1 eV can be used to change the overall electronic properties.[46] The material's high oxidation potential makes it difficult for PT polymerization and deposition on large insulating surfaces. Various techniques such as oxidative chemical vapor deposition, electrochemical oxidation and chemical oxidation in solution have been used to make polymeric thin films using thiophene monomers. Electrochemical oxidation is one among them. Because it is simple to manage the degree of polymerization, polymerization is the most widely used technology. Moreover, electrospinning can be used to form PT nanofiber architectures. Poly(3-alkylthiophene)s (P3AT) is one of the most significant classes of PTs, which exhibit good electrical and thermal conductivity, processability and environmental stability.[45] However, P3AT is expensive. Thus, there are just a few feasible uses for it. Reasonable balance between the solubility and electric properties of CPs like poly(3-hexylthiophene), poly(3-pentylthiophene) and poly(3-butylthiophene) makes them to utilize in organic electronic sensing applications. All of these substances belong to the P3AT family. It becomes possible to accomplish balanced hole/electron transport as the number of alkyl chains increases due to the continually increasing degree of phase separation.[45]

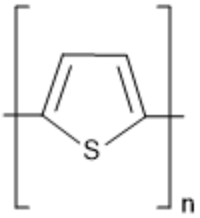

FIGURE 5: STRUCTURAL ILLUSTRATION OF POLYTHIOPHENE

FACTORS AFFECTING CONDUCTIVITY OF CONDUCTING POLYMERS

Conducting polymers, as their name suggests, have the capacity to conduct electricity related to a particular mechanism. Thus the decisive variable, conductance, is a key characteristic that makes it crucial to use in a variety of fields, such as energy storage. Temperature, solvent, doping, polymerization technique, etc. are significant factors in controlling the conductivity of electroactive materials, including conducting polymers.[47]

A. EFFECT OF POLYMERIZATION TECHNIQUE

For the synthesis of polymers from monomers, several polymerization procedures, including environmentally friendly ones, are used.

a. Chemical oxidation method

Lewis's acid, ammonium persulfate, and other oxidizing agents can oxidize the monomer and form cation and cation radical sites in it.[48] However, "head to head", "tail to tail", and "head to tail" type connections are rarely conceivable since they need the abstraction of two protons in order to make a covalent connection between monomer molecules.[49] Chain growth and polycondensation are the two techniques used in chemical oxidative polymerization. In contrast to chain development, which involves electrophilic substitution, polycondensation involves the recombination of cation radicle oxidation sites.

b. Electrochemical polymerization

A simple, safe and ecofriendly method to convert monomer into conducting polymer is electrochemical polymerization, sometimes referred to as electropolymerization. This technique makes use of oxidant over a working electrode which initiates polymerization. This green polymerization method is free of harmful chemicals and has high reproducibility and good electrical and electronic characteristics.[50] The two common mechanisms that include the oxidation of the monomer over the substrate are anodic and cathodic electropolymerization. The coupling of neighboring cations or interaction between neutral monomer or radical cation are two possible mechanisms of polymerization.

c. Vapor phase polymerization

This method makes use of vapor-phased monomer to a substrate to initiate the polymerization. The lack of solvent, the fact that monomer is a vapor component, which reduces the chance of agglomeration and moreover, the conductivity of the conducting polymer produced in this polymerization is high are the benefits of vapor phase polymerization than other methods.[51] This process makes it feasible to manufacture electrochemical energy storage devices, capacitors and organic solar cells.

d. Photochemical method

Conducting polymers can also be made by exposing them to light and ultraviolet rays in the presence of a catalyst. According to the mechanism, there are two different kinds of photochemical reactions: photopolymerization in the presence of an appropriate catalyst and monomer excitation in the presence of light. Lengthy reaction time and low quantum efficiency makes the second approach rarely applied .[52]

B. EFFECT OF DOPANT

Proton donors, long-chain substances like nitrosyl sulfuric acid (NSA), camphor sulfonic acid (CSA), dodecyl benzene sulfonic acid (DBSA) and p-type (oxidative process) and n-type (reductive process) dopants can all be used to dope organic compounds to increase their electrical conductivity and solubility.[53] While p-type doping involves the oxidation of polymers by dopants and leaves them with a positive charge, n-type doping involves the reduction of polymers by dopants and leaves them with a negative charge on them. These dopants have the ability to enhance conductivity as well as electrical properties. Dopants with polarons and bipolarons, two different types of charge carriers, can be used to provide conducting character in conducting polymers as shown in Figure (6).[54] The size, type and quantity of dopants used to dope polymers determine their conductivity. When choosing a dopant method, the size of the dopants being employed for doping purposes is a key consideration. The size of the dopants, which includes small dopants like Br, Cl, etc. as well as larger dopants like p-Toluene sulfonic acid (PTSA), dodecyl benzene sulfonic acid (DBSA) etc. affects the structural characteristics and conductivity of conducting polymers. [55] The conductivity of doped materials increases up to a certain point (maximum conductivity at a charge carrier density of 10^{18}–10^{21}), but after that point, a downward trend appears that can be related to a change in the microstructure of the polymer.[56] Since doping is reversible, dedoping can result in polymers having semiconductive characteristics.[47] There are several doping techniques used, including self-doping, neutron irradiation, electrochemical doping, gaseous doping, radiation-induced doping, and others.[57]

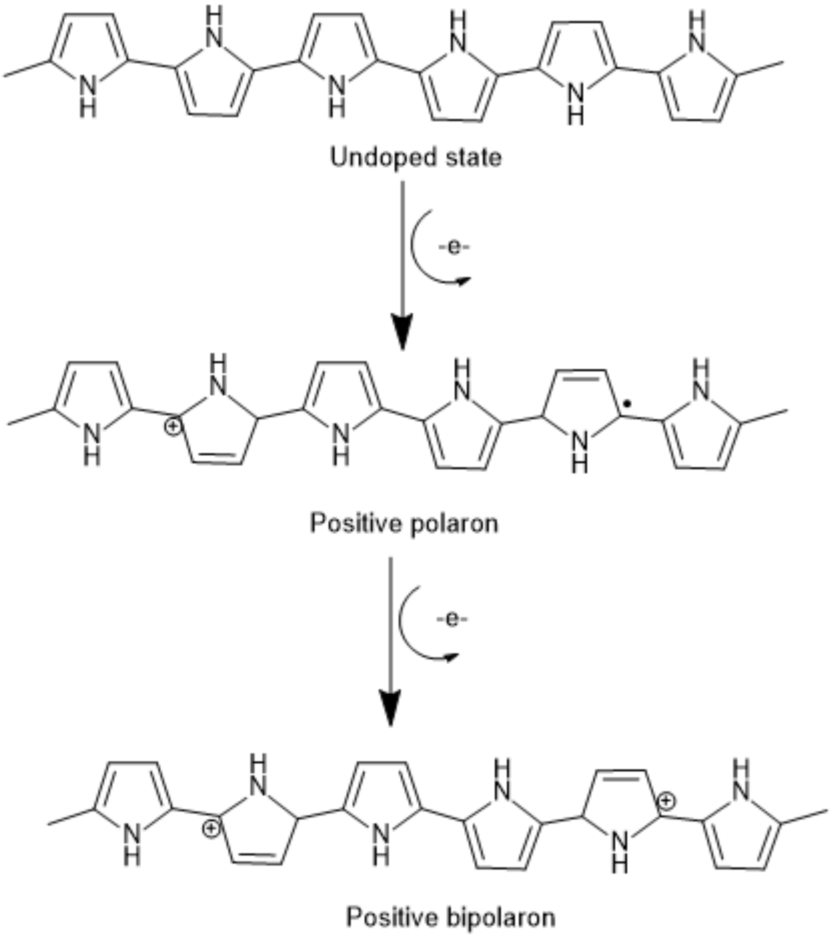

FIGURE 6: CHEMICAL STRUCTURES OF POLYPYRROLE POLARON AND BIPOLARON

C. EFFECT OF TEMPERATURE

Conducting polymers' electrical conductivity is strongly influenced by temperature; as temperature rises, the material's resistivity significantly decreases as a result of the massive emission of free electrons (charge carriers), which causes high thermal excitation and increased electronic conduction.[47] Study shows that when temperature rises, limited conductivity changes occurs in samples that are heavily doped, but significant alteration occur in samples that have just moderately or lightly doped.[58] Arrhenius plots and Mott plots are frequently studied for a deeper knowledge of the electrical conductivity of conducting polymers. Equation 1 (the Arrhenius equation) is used to represent the interaction of charge carriers near room temperature. The Mott equation (Equation 2), which may be produced, can be used to analyse the overlapping wave functions of neighbouring localised charge centres in the Mott plot.[59] The temperature is shown to have a significant impact on or be a function of conductivity of conducting polymer.

Arrhenius equation:

$$\sigma = \sigma_O \exp\left(-\frac{E_a}{kT}\right) \qquad (1)$$

where σ is conductivity, E_a is activation energy, σ_O is DC conductivity, k is Boltzmann constant and T is absolute temperature.

Mott equation:

$$\sigma = K_0 T^{-1/2} \exp\left[-\left(\frac{T_O}{T}\right)^{1/4}\right] \qquad (2)$$

where σ is conductivity, K_o is Mott characteristic conductivity parameter, T is absolute temperature, T_o is Mott characteristic temperature.

According to Yamada *et al.*, the temperature drops results in decrease in charge carrier density.[60] Camphor sulfonate doped PPy film prepared electrochemically at varying temperatures for supercapacitor indicates that conductivity drops as the temperature increases (10-30˚C being optimum temperature).

D. EFFECT OF BANDGAP

A distinguishing feature of conducting materials (metals or semiconductors) is the bandgap, which is the energy (Eg) required to excite an electron from the conduction band (CB) to the valence band (VB) in inorganic semiconductors or from the highest to the lowest occupied molecular orbital (HOMO to LUMO) in organic semiconductors. [43] [61-64] The conductivity of conducting polymer is strongly influenced by the band gap; the larger the band gap, the lower is the conductivity. In conducting polymers, variations in the bond length may be utilized as a predictor of the bandgap; a bigger bandgap is caused by a greater variation in the C-C bond length, which results in less conductivity. In addition to the bond length change, additional elements in Figure (7), include planarity, the resonance energy of the conjugated structure, π - conjugation length, the presence of substituents and their effects, the dihedral angle (θ), intermolecular interactions, etc. [65] Conducting polymers are typically large bandgap insulators that have further processing to become conductors by oxidation or reduction. According to Reyes et al., the conductivity rises as the size of the non-substituted oligomer furan increases and the difference between HOMO and LUMO decreases. Additionally, substituted oligomers have lower bandgap energies than unsubstituted oligomers, thus polyfuran can exist in dicationic and dianioninc forms (0.15 and 0.17 eV) respectively.

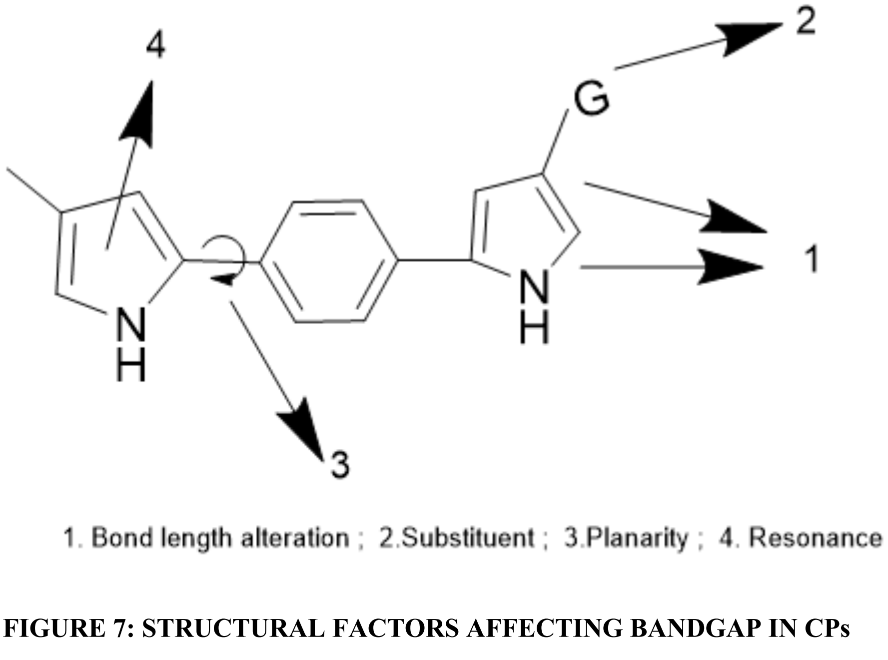

FIGURE 7: STRUCTURAL FACTORS AFFECTING BANDGAP IN CPs

E. EFFECT OF EXTENDED CONJUGATION

With conducting polymers, the degree of conjugation directly relates to electrical conductivity (poor electrical conductivity is caused by low π conjugation). [66] Conducting polymers, particularly those with small band gaps and extended conjugation, are used in semiconductors because of their optical and electrical characteristics. An organic polymer with a conjugated system named PA can develop a high electrical conductivity (17000 S/cm) when doped with iodine. According to the studies, the electrical conductivity of trans-PA drops by eight times when the conjugation length is reduced from 100 Å to 10Å.

F. EFFECT OF FUNCTIONAL GROUPS

The kind of functional groups, such as electron withdrawing groups (EWG) or electron donating groups (EDG), which are present in the molecule affects the redox potential, stability, and conductance of conducting polymers. Depending on the position, number, and type of substituents, the steric and electronic effects of functional groups on conducting polymers modify their electrical, electrochemical, and a variety of other physical properties. The redox properties of PANI are significantly influenced by the type of functional groups and steric or electronic effects. By lowering the electron density on the nitrogen atom, the EDG increases the oxidation potential as opposed to the EWG, which reduces it. EDG, such as -CH_3, was discovered to decrease steric strain by increasing the torsion angle between the conjugated rings with increased distance between the polymer units.[67]

APPLICATIONS OF CONDUCTING POLYMERS

Conducting polymers can be used in a variety of applications due to their advantageous electrochemical behavior, high theoretical capacitance, robust redox activity, excellent wave absorption, and electrical properties. A few are explained here:

A. SUPERCAPACITORS

Supercapacitors have attracted tremendous commercial attention because of growing markets for electronic automobiles, wearable gadgets, etc. The key difference between supercapacitor technology and conventional capacitors is that the former can store 1000 times more energy. In addition, they include high energy and power densities, high speed charge discharge cycle and outstanding cycle life.[68] There are three types of supercapacitors based on how they store their charge. The contact between the electrode and electrolyte in an electric double layer capacitor (EDLC) is where the charge accumulates and the device operates with the usage of non- faradic technique. Capacitance is produced by an electrochemical process that results in the development of a double layer of charge. Pseudocapacitors store charge via a faradic method that uses a redox reaction or intercalation. Pseudocapacitors make use of the redox activity of conducing polymers [69] and transition metal oxide such as Co_3O_4 [70], Mn_3O_4 [71] as electrodes. EDLCs and pseudocapacitors combination have been used in hybrid supercapacitors. Due to their intrinsically flexible polymeric nature, conducting polymers are the most potential active electrode when compared to the conventional active electrode materials, making them perfect for supercapacitor application.[72] A variety of morphologies, including particles, rod-like structures, sheets and hydrogels, are obtainable for conducting polymers. [73]

On a transparent indium tin oxide (ITO) substrate, Kulia *et al.* constructed a sequential array of vertically aligned PANI nanorods using a nanotemplate based on supramolecular assemblies of block copolymers. The greatest capacitance of the nano-spaced PANI nanorod arrays, which had outstanding electrochemical characteristics, was 3407F/g.[74] Up to 500 cycles the PANI nanorods exhibit high electrochemical stability, but beyond that, they act irregularly and do not exhibit a regular charge-discharge cycle. This may be due to the less stability of small dimension PANI nanorods. Through the use of galvanostatic current, Wang Kai produced an extensive series of aligned PANI nanowires. [75] Huge arrays of nanowires with narrow diameters that were positioned perpendicular to the substrate when PANI was used as the supercapacitor electrode assisted in ion diffusion, as seen in Figure (8). PANI nanowire arrays' maximum specific capacitance, which was measured at 950 F/g, was recorded. Enhanced electrochemical performance of functional nanomaterials could be observed by the orientation of nanostructures. [75]

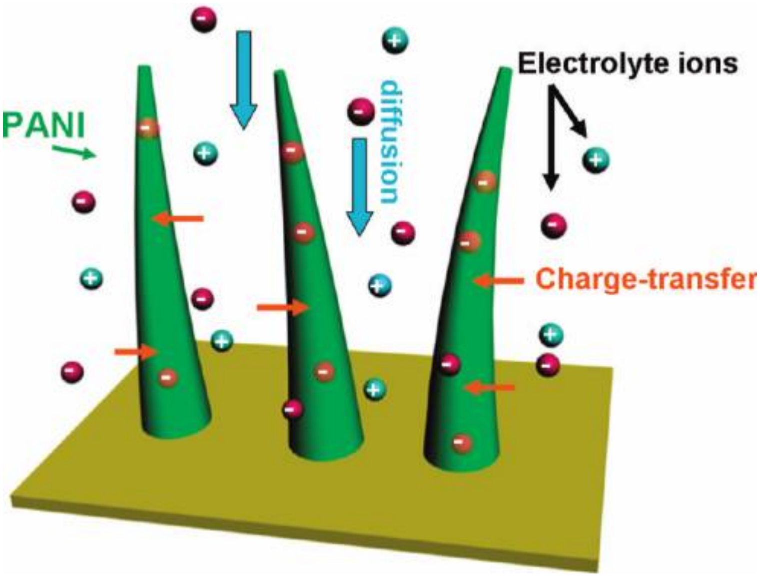

FIGURE 8: SCHEMATIC ILLUSTRATION OF THE OPTIMISED ION DIFFUSION PATH IN NANOWIRE ARRAYS. Reprinted with the permission from [75,] copyright 2010, American Chemistry Society.

As illustrated in Figure (9), Afsaneh *et al.* fabricated a high-performance asymmetric supercapacitor using a novel ternary binder-free nanocomposite (PPy/FeCoS-rGO/NF) composed of polypyrrole, iron-cobalt sulfide, and reduced graphene oxide on nickel foam electrode. In addition to improving the uniform growth of conductive PPy film and FeCoS ultrathin porous nanosheet, 3D porous graphene foam (GF) also significantly improves mechanical stability, rate capability and energy storage capacity. The PPy/FeCoSrGO/NF exhibits outstanding electrochemical properties, having a high specific capacitance of 3178F/g at 2A/g and an acceptable rate capability. High surface area, excellent electronic conductivity, low contact resistance between the PPy/FeCoS-rGO active layer and the Ni foam current collector, a short electrolyte ion diffusion pathway, quick electron transfers, and effective use of active material during Faradic charge-storage processes are all components of the excellent electrochemical performance. Taking advantage of their superior properties, nickel foam covered with reduced graphene oxide (rGO/NF) is used as the negative electrode and PPy/FeCoS-rGO/NF is used as the positive electrode to produce a hybrid asymmetric supercapacitor. The specific capacitance of a device made up of PPy/FeCoSrGO/rGO is high, measuring 94 F/g at 1 A/g. The asymmetric supercapacitor device has outstanding cycling performance with 97.5% capacitance retention after 2500 cycles.[76]

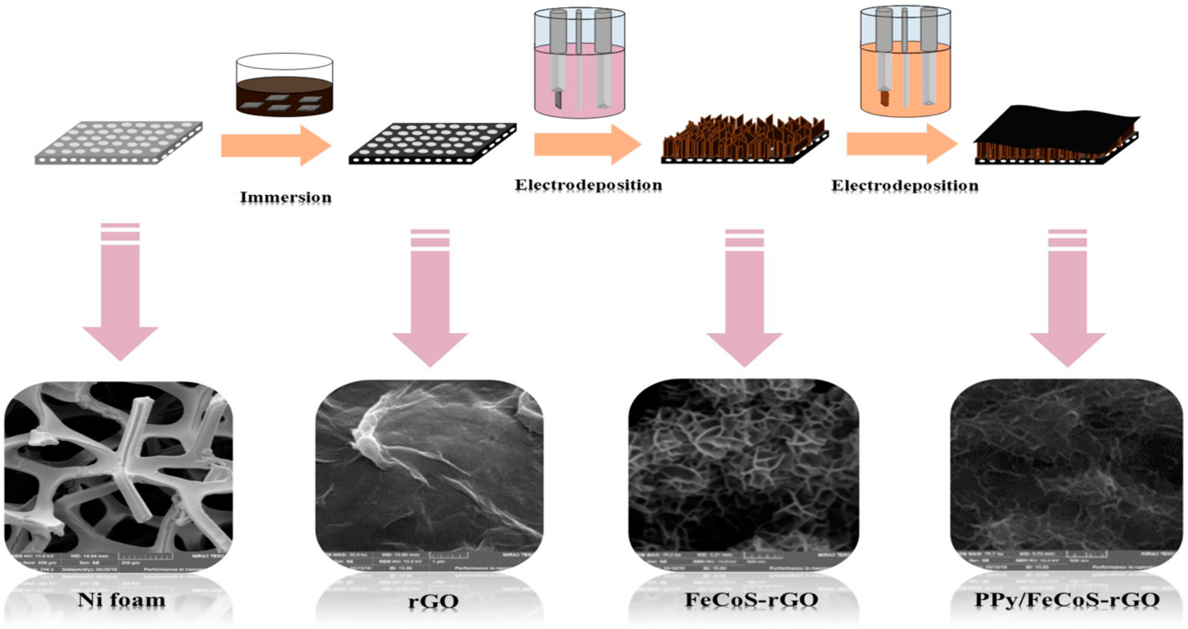

FIGURE 9: SCHEMATIC ILLUSTRATION OF PREPARATION OF PPy/FeCoS- rGO ELECTRODE ON Ni FOAM SUBSTRATE. Reprinted with the permission from [76,] copyright 2020, American Chemistry Society.

Anthony *et al.* demonstrated a simple drop-cast of a commercial PEDOT: PSS solution on 3 nm alumina coated silicon nanowires, a quick and easy approach for depositing a very reliable pseudocapacitive substance as shown in Figure (10). Combining epitaxially grown, highly doped silicon nanowires with an alumina atomic layer deposition (ALD) coated layer for protection and embedding them in a gelatinous-like polymer matrix was feasible. The composite material generated by PEDOT: PSS-Alumina coated Si Nanowires (PPSS-A@ SiNWs) demonstrated excellent capacitive behaviour with a specific capacitance of 3.4 mF/cm^2 at a current density of 2 A/g in an aqueous Na_2SO_4 electrolyte. With more than 500 000 cycles at a current density of 0.5 A/g, micro supercapacitor (MSC) devices based on this material exhibit excellent lifetime capacity, maintaining 95% of their initial capacitance. [77]

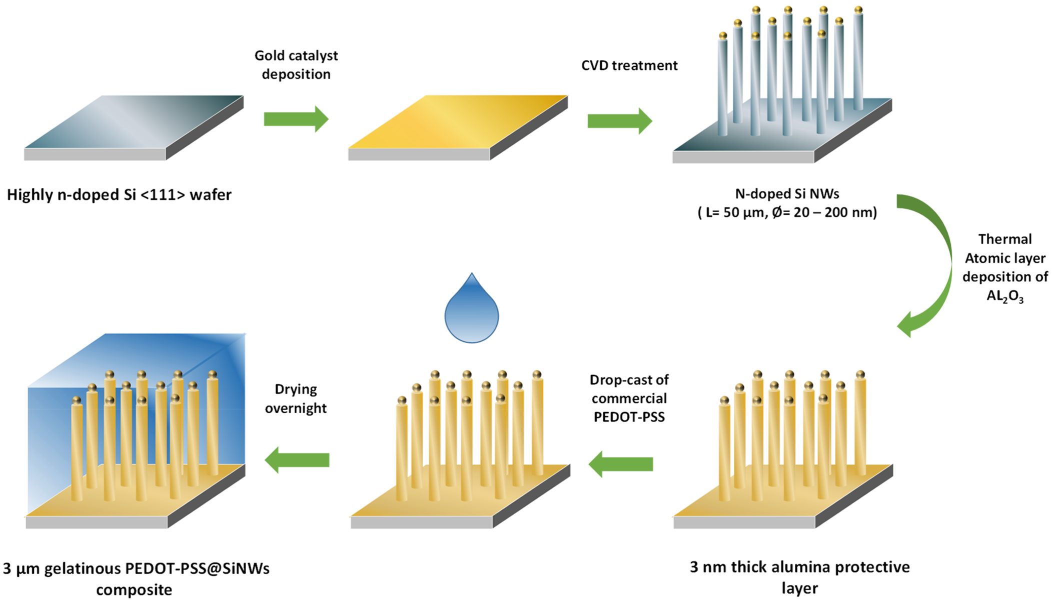

FIGURE 10: SCHEMATIC ILLUSTRATION OF MICRO SUPERCAPACITOR ELECTRODE MANUFACTURING INCLUDING CHEMICAL VAPOUR DEPOSITION, ATOMIC LAYER DEPOSITION AND DROP CAST. Reprinted with the permission from [77] copyright 2018, American Chemistry Society.

B. CORROSION INHIBITION

Metals are anticipated to be employed in the construction, automobiles, energy, aerospace, home sectors, etc. Metals rapidly deteriorate when exposed to moisture, acid or alkaline or both environments. Corrosion inhibition techniques and coatings have been attributed to preventing the attack on them. Earlier cathodic and anodic inhibitors make use of chromate coatings. Because they are carcinogenic and dangerous to both human life and the environment, chromates and coatings containing chromium have a major drawback. Corrosion prevention is greatly improved by conducting polymers and coatings that contain conducting polymer pigments. The ability of conducting polymers to preserve the continuous passivity of metal during anodizing and O_2 reduction on the film's surface has attracted growing interest as a potential component of corrosion-resistant coating systems. Between metal and coating, a metal oxide interface layer is formed. Listed below are the processes behind polymers' corrosion inhibition property:

a. An electric field is produced when conducting polymer and metal comes in contact. The movement of electrons from metal to oxidized species is constrained by this electric field. As a result, corrosion is inhibited.[78]

b. A metal oxide layer can be formed by polyaniline in the anodic protection mechanism, both above and below the metal surface. The metal oxide layer serves as a major source for corrosion inhibition.[79]

c. Conducting polymers, particularly polyaniline, have the controlled release of inhibitors during reduction. When the reduction takes place, an anion will be released from a conducting polymer based coating that has been applied to the metal surface. This helps in the reduction of corrosion.[80]

Sathyanarayanan *et al.* demonstrated vinyl acrylic organic and polyaniline pigmented epoxy. Its performance has been evaluated on stainless steel. Impedance spectroscopy first decreases due to the pinholes generated on the metal surface, then increases as exposure duration rises. An interface layer of metal oxide between the metal surface and the coating below was absorbed by the coating as the exposure period increased.[81] Thus, it was determined that the exposure period and coating thickness affect how efficient polyaniline pigmented organic coatings are in preventing corrosion.

Armelin *et al.* examined polyaniline and polypyrrole individually for their effectiveness as anticorrosive additives in zinc epoxy coating. By exhibiting reversible redox properties akin to chromate coatings, it demonstrates that the corrosion prevention property was enhanced. Zinc pigments form zinc oxide, which defends the metal layer and reduces the likelihood of corrosion. Additionally, the study showed that polypyrrole percentage above a particular threshold cannot demonstrate superior inhibitory properties.[82] Chen Fei and Liu Peng in Figure (11) depicts a new technique to develop coatings containing the polyaniline (PANI) compound that resist corrosion in aqueous environments.[83] First, via chemical oxidative dispersion polymerization in the presence of partially phosphorylated poly (vinyl alcohol) (P-PVA), conducting polyaniline/partially phosphorylated poly (vinyl alcohol) (PANI/P-PVA) spherical nanoparticles with considerable water dispersibility were generated. The aqueous PANI/P-PVA coatings were created and applied to mild steel using a dipping technique. To evaluate the corrosion resistance of PANI/P-PVA-containing coatings on mild steel, a salt spray test and electrochemical impedance spectroscopy (EIS) method were utilized in a 3.0 wt% sodium chloride aqueous solution. The effective Fe_2O_3 passive layer and uniform distribution of the system allowed the waterborne PANI/P-PVA-containing coatings to offer higher corrosion protection than the PANI ES-containing coatings with a PANI ES content of 2.5 wt%, as demonstrated by alteration of the open circuit potential (OCP) values and EIS spectra with exposure time.[83]

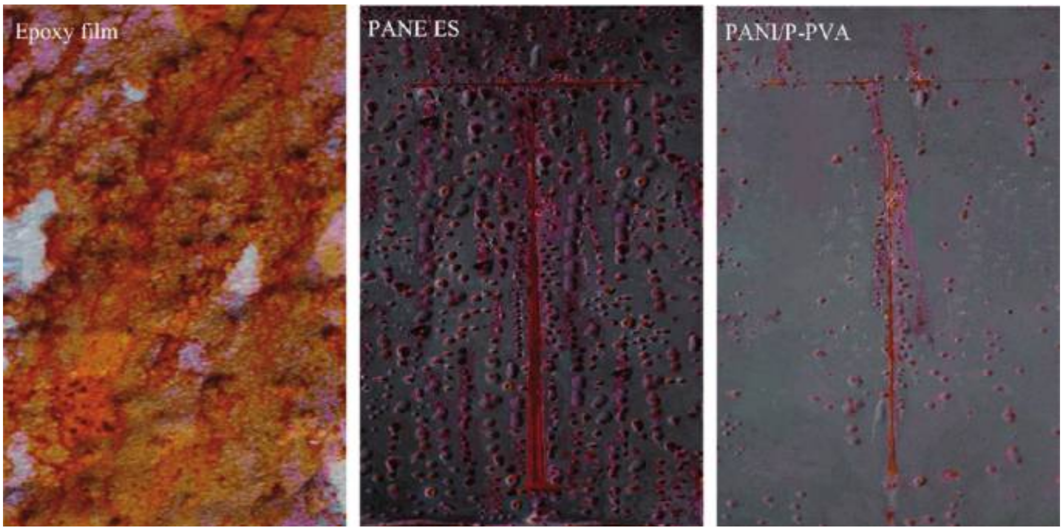

FIGURE 11: PHOTOGRAPHY OF PURE EPOXY COATED PANEL, 2.5 WT % PANI ES-COATED PANEL AND 2.5 WT % PANI/P-PVA-COATED PANEL AFTER 30 DAYS EXPOSURE TO SALT SPRAY TEST. Reprinted with the permission from [83]copyright 2011, American Chemistry Society.

The PANI/TiO_2 coating demonstrated high thermostability, anticorrosive and hydrophobic characteristics. Heat dissipation increases by thermal conductivity; hence thermal conductivity plays an important role in durability. It prevents thermal expansion-related coating delamination. The contact angle of PANI/TiO_2 was 150˚, which clearly indicates the super-hydrophobicity of the material.[84]

C. GAS SENSORS

Gases produced by industrial and natural processes need to be recognized and analyzed due to their considerable impact on human health. Due to its advantages, such as adjustable conductivity (from insulator to metallic conductivity), low density, room temperature operation, affordability, environmental stability and many functionalization options, gas sensors made from conducting polymer sensing materials have generated interest. Gaseous ammonia (NH_3) is a prominent air contaminant. Although it is safe in modest doses, large amounts may lead to the destruction of the mucosa in the respiratory tract, eye and skin irritation and even death. Animal husbandry, production of urea for agricultural use, automobile exhaust and industrial manufacture are all sources of ammonia in the air. Polypyrrole and polyaniline are the conducting polymers that are most frequently used for gas sensors whose mechanism is shown in Figure (12).[85]

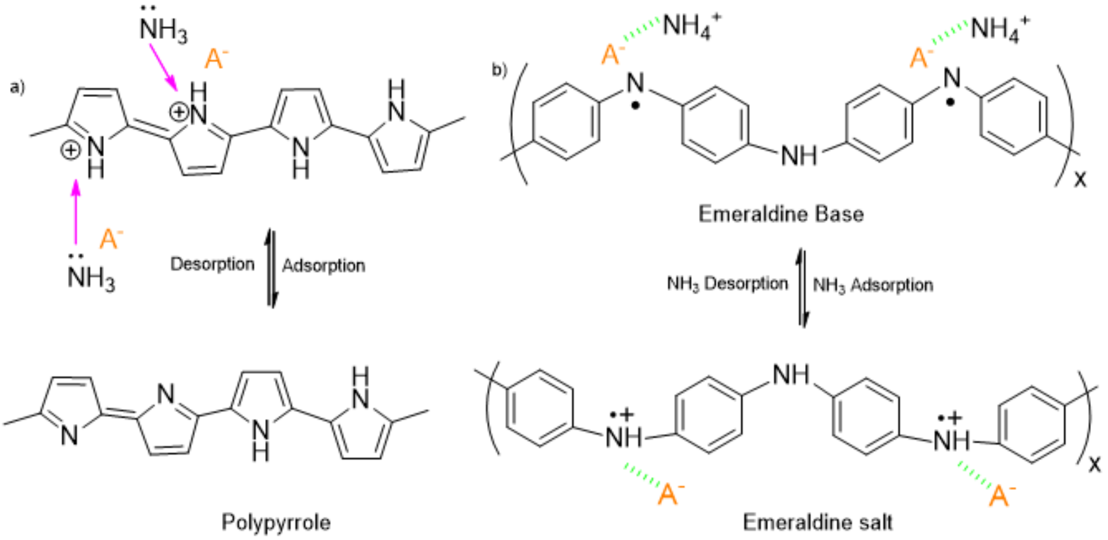

FIGURE 12: AMMONIA SENSING MECHANISM BY a) PPy b) PANI BASED GAS SENSOR

She Changkun *et al.* manufactured polypyrrole based gas sensor involving the comparison of two flexible substrates, sponge and silk-fiber.[86] The PPy film was synthesized by in-situ chemical oxidation using silica nanospheres (NS) as a template. The response from the PPy/NS@ silk fiber sensor (73.25%) was five times greater than that from the PPy/NS@ sponge sensor (14.51%) when subjected to 100 ppm NH3 at (68 ± 5%) relative humidity at ambient temperature. Additionally, the silk fibers offer a solid foundation for the attachment PPy, and the PPy/Ns@ silk fiber sensor has more active sites to deliver a strong dynamic response and high sensing response.[86] Lalit *et al.* fabricated flexible PANI -polyethylene terephthalate (PET) ammonia sensors through in-situ polymerization processes at various temperatures. At room temperature, this flexible PANI sensor displayed high stability, good responsiveness, reproducibility and low detection limit (5ppm) to ammonia vapors. The sensor's resistance almost remains the same after repeated bending and its sensing capability is unaffected, indicating the sensor's mechanical stability and flexibility.[87]

The dual role of PANI in boosting flexibility and ammonia sensing performance was examined by Maity Debasis *et al.* [88]. Spray-coated MWCNTs on polypropylene woven fabric were used to produce the wearable, which was then chemically polymerized via in-situ polymerization in Figure (13). The sensor changed its resistance in response to ammonia vapors extremely fast (almost 9 seconds), and it swiftly returned to its baseline resistance when ammonia was not present (recovery time: 30 seconds). In the presence of potential cross-interfering gases, MWCNTs/PANI sensors have been demonstrated to be particularly sensitive to ammonia. After a month of monitoring, the sensor has displayed outstanding sensing stability. [88]

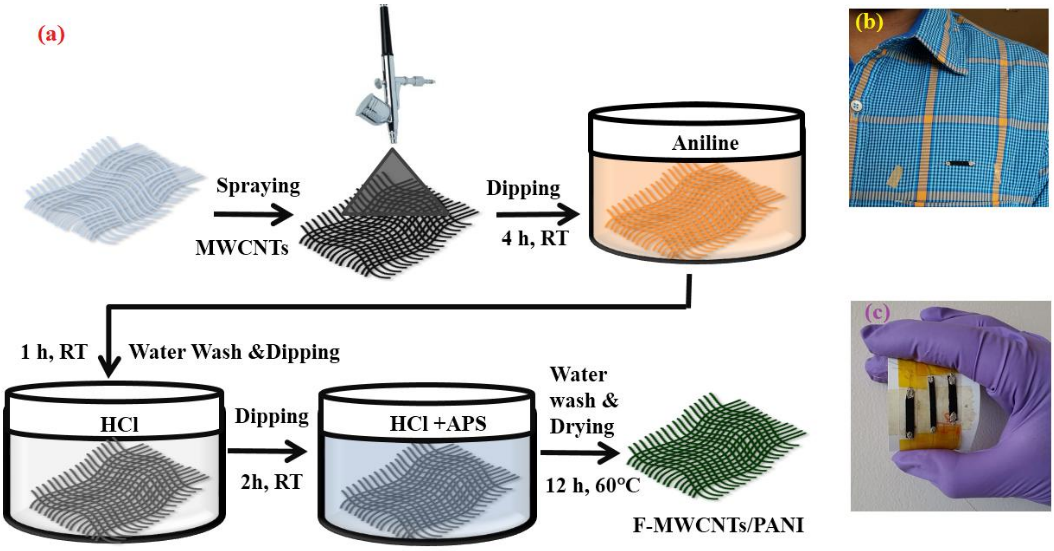

FIGURE 13: a) SCHEMATIC SHOWING SPRAY DEPOSITION OF MWCNTs ON THE (F-MWCNTs) AND PANI SYNTHESIS ON MWCNTs COATED FABRIC SURFACE, b) SENSOR INTEGRATION IN SHIRT AND c) SENSOR ARRAY FOR AMMONIA SENSING. Reprinted with the permission from [88] copyright 2018, American Chemistry Society.

He Yuan *et al.* described the manufacturing of conductive polymer coatings and patterns using an oxidant-intermediated surface polymerization (OISP) process on a variety of substrates. On a polyethylene terephthalate (PET) substrate, a coating or pattern composed of densely packed colloidal vanadium oxide (V_2O_5-nH_2O) nanowires was applied by spin coating, dip coating, or printing. Then, using in situ oxidation polymerization, this coating or pattern is converted into a conducting polymer. It is suitable for producing sensor arrays because the coating approach provides a simple way of integrating a variety of CP patterns on a compact substrate. The same PET substrate was used to print three V_2O_5-nH_2O patterns, which were afterward transformed into CP such as PPY, PANI and PEDOT as in figure (14A). Each CP design was turned into a resistor-type sensor after carbon electrodes were printed. Each sensor's signal amplitude S varied significantly because each analyte and each CP interacted differently. Three channels of relative resistance change signals ($S = \Delta R/R$) were gathered as a consequence of each sensor's resistance changing when analyte gas was supplied. As a result of the unique interactions between each CP and each analyte, each sensor's signal amplitude S differed significantly from the others. If the three signals SPPY, SPEDOT, and SPANI are merged to form two new variables, P1 = SPPY/SPEDOT, and P2 = SPPY/SPANI, the points (P1 and P2) corresponding to the various analytes will be located in the various areas of the coordinate system. Figure (14B) demonstrates how sensor array allowed to discriminate between the vapors of ammonia, methanol and n-hexane based on their positions in the P1-P2 plot. The region was relatively independent of the analyte concentration. The aforementioned cross-reactive sensor array therefore assists in identifying an unknown sample that may be one of the three vapors indicated above by utilizing the locations of the points (P1 and P2) in the plot. [89]

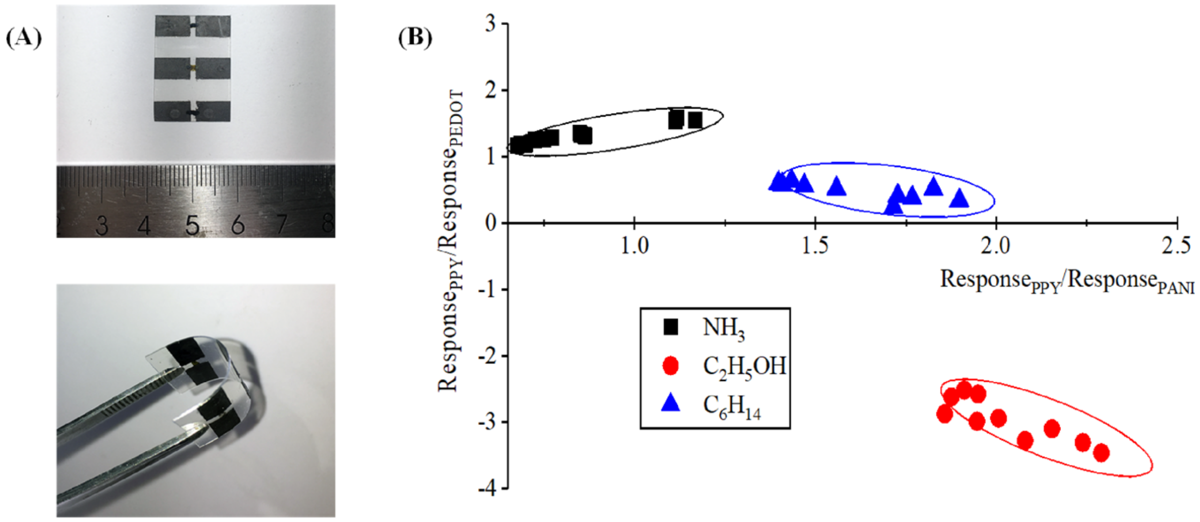

FIGURE 14: GAS SENSING PERFORMANCE OF A FLEXIBLE SENSOR ARRAY FABRICATED WITH OISP (A) PHOTOGRAPHS OF FLEXIBLE SENSOR ARRAY MADE FROM PPy, PANI AND PEDOT FILMS. (B) PERFORMANCE OF THE SENSOR ARRAY IN DISTINGUISHING DIFFERENT GASES. Reprinted with the permission from [89]copyright 2021, American Chemistry Society.

D. CONDUCTING POLYMER HYDROGELS (CPH) FOR BIOELECTRONICS

Hydrogels and conducting polymer are combined to produce conducting polymer hydrogels, which have unique features that are not possible with traditional materials. The CPH are composed of hydrophilic networks that are covalently or physically cross-linked with conducting polymers like polyaniline, polypyrrole, or polythiophene.[90] High self-healing, electrical, mechanical and biocompatibility are characteristics of CPH. Tissue engineering and bioelectronics make extensive use of it. Conducting polymer is inadequate for certain applications and is substituted by CPH due to its long-term stability. Various techniques, such as Michael-type addition polymerization, click chemistry and free radicals, are utilized for the manufacturing of CPH. Human's reasoning ability, muscular motion and thinking are all made possible by the electronic response of the nervous system. Electrodes that interact with biological tissues make up bioelectronics interfaces. They gather bioelectronic signals and provide customized tissue self-monitoring.[91] The use of bioelectronics has been widespread in many fields, including electronic skin, wearable, flexible implanted devices and the monitoring of brain activity.[92]

Ren Xiaoning *et al*. utilising a straightforward solution-mixing procedure for synthesizing highly conductive PPy-PEDOT: PSS hybrid hydrogels. [93]. In this manufacturing approach, pyrrole monomer and PEDOT: PSS dispersion were combined, and in-situ chemical oxidative polymerization was used to yield polypyrrole (PPy). Hybrid PPy-PEDOT: PSS hydrogels have been produced by the electrostatic interaction of negatively charged PSS and positively charged conjugated PPy. The hybrid PPy-PEDOT: PSS hydrogels exhibit a conductivity of 867 S/m. High biocompatibility is demonstrated by the hybrid hydrogels of PPy-PEDOT: PSS. The hydrogels in Figure (15) can more easily support 3D cell growth due to the hierarchical porosity of the PPy-PEDOT: PSS hybrid hydrogels. The remarkable in situ biomolecular detection and real-time cell proliferation monitoring abilities of the PPy-PEDOT: PSS hybrid hydrogels suggest their potential as incredibly sensitive electrochemical biosensors for bioelectronics applications.[93]

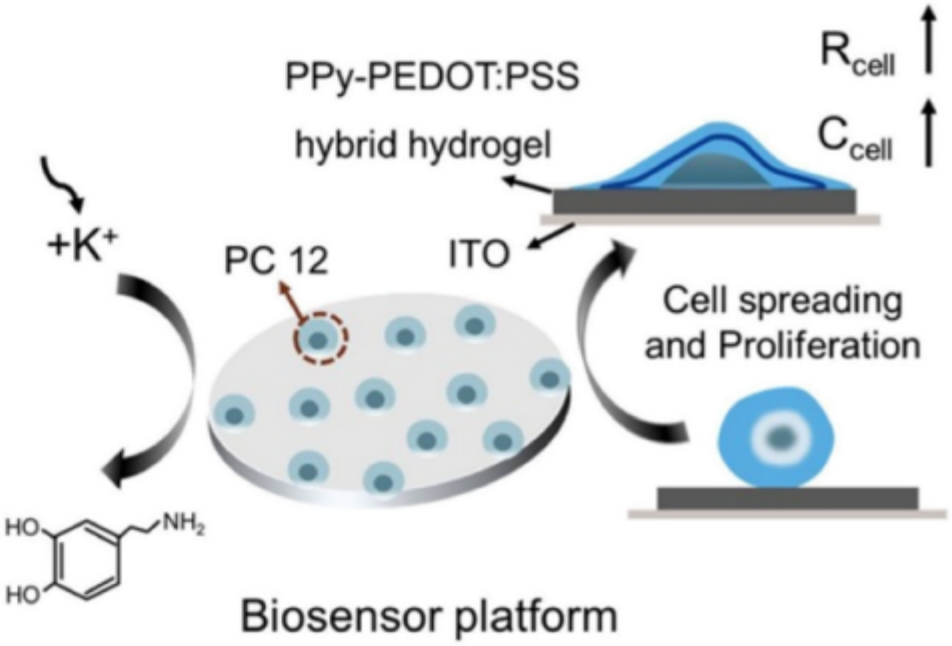

FIGURE 15: REAL-TIME MONITORING AND IN-SITU SENSING OF LIVING PC12 CELLS USING PPY-PEDOT: PSSHYBRID HYDROGEL BASED BIOSENSORS. Reprinted with the permission from [93] copyright 2021, American Chemistry Society.

As a result of its ability to transform mechanical impulses into electrical signals, CPH are used for developing pressure and strain sensors. To monitor electrophysiological signals, such as electromyography, electroencephalography, and ECG, flexible electrodes based on CPH are often used. Sometimes extremely weak electrophysiological signals are detected. Conductivity and signal-to-noise ratio improvements are the device's key fabrication problems. Additional factors to be taken into account include biocompatibility, long-term stability, and skin adherence. Hydrogels made of PAA/PEDOT have been created to recognize a range of electrophysiological signals. Real-time monitoring of lactate and glucose is done with hydrogels based on polyaniline.[94]

Conductive fillers can be added to boost their conductivity. Chen Po-Yen et al. developed a facile phytic acid mediated synthesis technique for PANI CPH, which shows a conductivity of 0.21S/cm.[95] . Multi-walled carbon nanotubes (MWNTs) were added, which allowed the MWNT-phytic acid complexes to form in a uniformly distributed state. The production of MWNT-PANI composite hydrogels was favored by the polymerization of PANI on the surface of MWNT. This results in increasing conductivity to 1.54S/cm. [95] The hydrogels exhibit phase separation between the polymer matrix and filler if the filler loading exceeds a certain level which results in reduced mechanical stretchability. Plasticizer can be included into the polymer matrix to address this defect. Solvent annealing of the conducting polymer PEDOT: PSS enhanced the stretchability by 35% and changed the interconnectivity of the fibrillary structure.[96]

E. BIOMEDICAL AND ANTIMICROBIAL APPLICATION

The removal of cancer cells from the human body may be accomplished by near-infrared (NIR) photothermal therapy (PTT). Fiber optics are used to light the target region. The use of photothermal materials to kill cancer cells appears to be less invasive when compared to other kinds of treatment like chemotherapy or radiation. PTT's targeted action, above other therapies, is one of its key benefits. This method reduces tissue loss around the active sites. As photothermal materials for PTT, materials with a high level of photostability, good NIR absorption, nanoscale dimension, low toxicity and less photocorrosion have a lot of possibilities. Figure (16) shows the excellent IR absorption capabilities and greatly reduced cytotoxicity of conducting polymers like polypyrrole, PEDOT, and PANI. Mei Chen et al. examined the control development of the 4TI tumour model using polypyrrole as a photothermal material. [97]. An experimental study using in vivo and in vitro assessments found that polypyrrole had a photothermal efficiency of 44.7% and outstanding cancer cell ablation capacity when exposed to 1W/cm^2 of NIR light. All irradiation tumors vanished after 60 days of treatment and no tumor development was recorded when the tumor size was evaluated at regular intervals.

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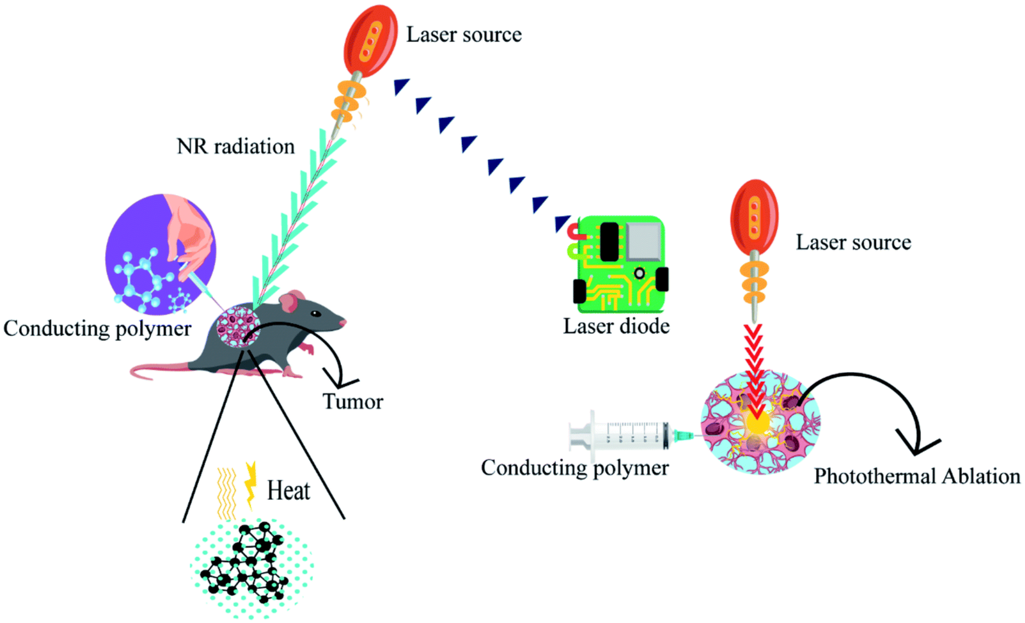

FIGURE 16: PHOTOTHERMAL TREATMENT SCHEMATIC REPRESENTATION USING CONDUCTING POLYMER. Reprinted with the permission from [18] copyright 2021, Royal Society of Chemistry.

Synthesized polyaniline composites have a potential future in photothermal treatment owing to their strong NIR absorption, superior water dispersion, ideal size, and high photothermal efficiency of 48.5%. Zero toxicity is a vital quality that supports in vivo research. Graphene, graphene oxide, and reduced graphene oxide are examples of carbonaceous materials that make good PTT materials because of their high surface area, NIR absorption, and efficient heat transfer. However, their use has been constrained by their poor dispersity.[98] High photothermal activity of polypyrrole is directed toward cancer cells. Chen and his coworkers investigated the photothermal efficiency of polypyrrole against 4T1 cancer cells without altering the material's surface. The biocompatibility of polypyrrole makes it possible for both in vivo and in vitro investigation. Due to its potent NIR absorption, research showed that polypyrrole has a 44.7% photothermal conversion efficiency and a high ablation efficiency. The antibacterial effect of conducting polymer materials suggests that they could perhaps be used in biomedical and other smart materials. They have the ability to increase the amount of H_2O_2, which encourages the generation of hydroxyl radicals and their subsequent antimicrobial actions. Studies have been done on PANI coated cloth that has been doped with phytic acid, which results in improved crosslinking between the PANI and the substrate. The bacterium and the polymer backbone interact electrostatically to provide the antibacterial activity. In Mueller-Hinton agar (MHA), which was incubated for 18 hours at 37 °C, Gram-positive Klebsiella pneumonia (K. pneumonia), Gram-negative Escherichia coli, and Gram-positive Staphylococcus aureus (S. aureus) were used to investigate the antibacterial activity of PANI-coated cotton and polyester fabric. The experiment demonstrates that cotton fabric exhibits no inhibition zone in the case of Escherichia coli (E. coli) or

Klebsiella pneumonia (K. pneumonia), whereas polyaniline coated polyester fabric has a superior inhibition zone of 10.3±0.04714.[99]

Several sectors such as the medical field, the food sector, the coating industry, etc., make use of conducting polymers as antimicrobial and antifungal agents. Against Campylobacter jejuni, Pseudomonas aeruginosa, Staphylococcus aureus and Escherichia coli, polyaniline and other conducting polymers show antibacterial properties. In Mueller-Hinton agar (MHA) plates, the efficiency of the gold @ PANI-itaconic acid-iron oxide composite material [Au@PANI-IA-Fe3O4] against Escherichia coli (Gram negative) and Staphylococcus aureus (Gram positive) germs has been examined. It has been discovered that the composite presents a minimal inhibition zone for E. coli bacteria but a more effective zone of inhibition for Staphylococcus aureus with increasing composite concentration. Candida albicans has been used as a test subject for antifungal property of composite. The outcome demonstrates that the composite exhibits a tendency towards increase in inhibitory zone when the composite concentration increases. [100]

F. AS AN ELECTRODE FOR ENERGY HARVESTING

One of the key characteristics of the material under consideration that helps the electrode materials withstand external forces and the risk of deformation is flexibility. Examples of CPs that can be used as electrodes in energy storage devices include PANI, PPY, PTH, and PEDOT due to their high conductivity, low cost, and exceptional mechanical flexibility, among other properties. Park Byung-Wook et al examined the usage of CP in dye sensing solar cells in Figure (17). [101] Conducting polymer poly(3,4-ethylenedioxythiophene) (PEDOT) doped with iron(III) tris-p-toluenesulfonate (PEDOT:Tos) with metallic conductivity was spin coated onto fluorine-doped tin oxide (FTO) glass and plain glass substrates for devices as a counter electrode (CE) in a dye-sensitized solar cell (DSC), with cobalt-based redox electrolyte. Impedance spectroscopy revealed that the charge-transfer resistance of the PEDOT:Tos-based counter electrodes in DSCs was five to ten times lower than that of the Pt/FTO CE. DSCs with PEDOT:Tos/glass CE obtained power conversion efficiencies (PCE) of 6.3%, which was comparable to DSCs with platinized FTO glass CE (6.1%). [101]

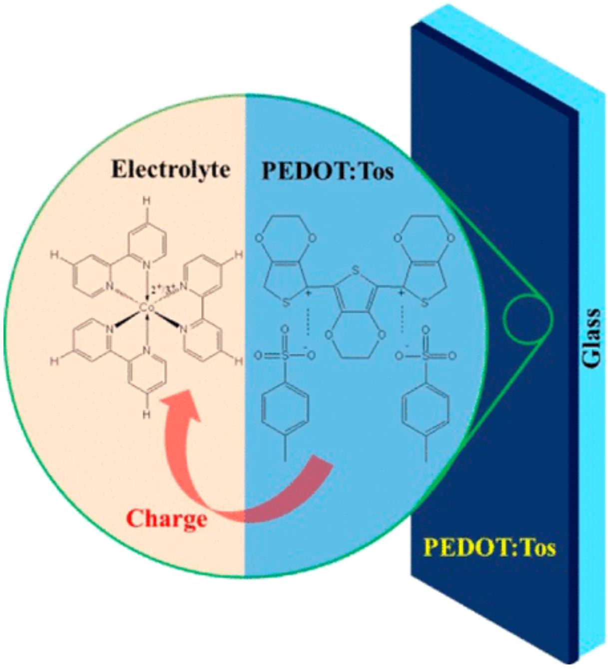

FIGURE 17: ILLUSTRATION OF DYE SENSITIZED SOLAR CELLUSING POLY(3,4-ETHYLENEDIOXYTHIOPHENE) DOPED WITH IRON(III) TRIS-P-TOLUENESULFONATE,
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CP can also be used for waste heat energy harvesting methodologies involving piezoelectric/ pyroelectric energy conversion. A piezo/pyroelectric device made entirely of polymers was constructed by Talemi Pejman et al. using beta phase PVDF as the active material and vapor phase polymerized (VPP) PEDOT as the flexible electrode overlay material. According to strain measurement up to the breaking-strain of PVDF (about 35%), the change in normalised sheet resistance R/R_o versus strain for the PEDOT-based electrodes was significantly lower than that of the platinum electrode. Measurements of the contact angle revealed that a higher energy surface had formed, which allowed for a more adhesive interaction between the PVDF film and the PEDOT coating. A mechanical cam actuator produced a measured piezoelectric output voltage that increased from 0.2 V to 0.5 V under the same strain conditions due to the improved contact between the two polymers. Pyroelectric open circuit voltage outputs of up to 100V and power outputs of up to 1μW (into a 60 MΩ resistive load) were generated by placing the film on and off of a hotplate with temperatures up to 50˚C above ambient.[102]

In our work, we focused on pyroelectric energy conversion (PEC) which could make use of CP as electrode for it. We synthesized PPy flexible electrode using chemical oxidation method which was coated on PET substrate. Along with that, PVDF which has high beta phase crystallinity is also been used for this PEC. The PPy film shows a resistance of 60 Ω/cm^2. The Fourier transform infrared spectroscopy (FTIR) in figure (18) shows that the band observed at 3227,2930,1655,1427,1070,923 cm^{-1} corresponds to N-H stretch, CH_2 group, C=C stretch, PPy ring stretch, C-H wagging, C-H in plane. Further studies are to done for the usage of PPy as electrode. [unpublished]

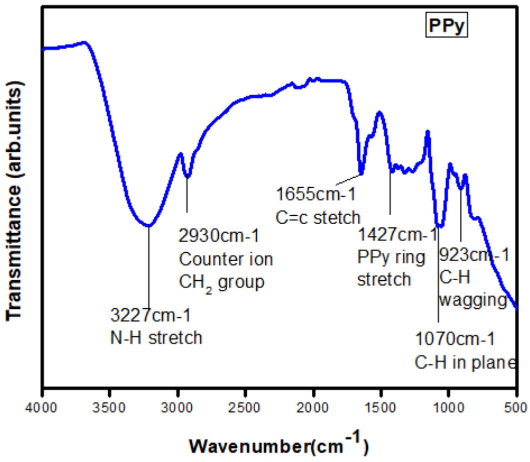

FIGURE 18: FTIR SPECTROSCOPY OF POLYPYRROLE.

CONCLUSION

Numerous outstanding qualities that mirror those of metals are seen for conducting polymers (CPs). Hence it attracted the attention of the scientific community, however some of these features have been shown to be superior to those of metals in specific applications. However, research based on CP used as electrode materials for flexible devices are not much reported, which enables attention in the research field. Hence CP are in limelight with various applications.

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