



Conducting Polymer Nanocomposites: Main Focus on Polyaniline-Based Photocatalysts

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Abstract

As a unique group of advanced polymer-based materials, conducting polymer nanocomposites combine the flexibility and conductivity of the polymer with the distinct properties of nanofillers. Conducting polymers have been widely investigated in various applications. Several conducting polymers, such as polyaniline (PANI), polypyrrole (PPy), poly (3,4- ethylenedioxythiophene) (PEDOT), and polythiophene (PTH) have been loaded with various semiconductor nanomaterials to prepare the composite photocatalysts. In this review, we summarized the synthesis, characterization, properties, and novel applications of conducting polymer nanocomposites especially photocatalytic degradation of hazardous chemicals. The synergic effect between conducting polymers and semiconductor nanomaterials were observed for a lot of composite photocatalysts.

Keywords: Conducting polymer nanocomposites, Polyaniline, Photocatalyst, Hybrid, Semiconductor nanomaterials

Introduction

Polymers were considered to be electrical insulators before the invention of conducting polymers (CPs) (conjugate polymers), but these organic polymers have unique electrical and optical properties similar to those of inorganic semiconductors. A conjugated carbon chain consists of alternating single and double bonds, where the highly delocalized, polarized, and electron-dense p bonds are responsible for its electrical and optical behavior. Typical CPs include polyacetylene (PA), polyaniline (PANI), polypyrrole (PPy), polythiophene (PTH), poly(para-phenylene) (PPP), poly(-phenylenevinylene) (PPV), and polyfuran (PF) (Figure 1) [1].

Conventional polymers consist of thousands to millions of monomer units. They are stiff and soluble in solvents, but a conjugated polymer chain consists of a lower number of monomer units. The mechanical property is gained by the presence of alternating single and double bonds present within it. The solubility and processability of CPs depend mainly upon the attached side chains, and the attached dopant ions give them mechanical, electrical and optical properties [2]. CPs are crystalline and partially amorphous. CPs consist of both localized and delocalized states, and the delocalization of p bonds depends heavily upon disorder, and this delocalization plays an essential role in the generation of charge carriers like polarons, bipolarons, solitons, etc., which are responsible for the transition from insulator to metal [3]. The conductivity of conjugate polymers acts like an insulator to a semiconductor in their pure form, and the conductivity increases with dopant concentration. In the undoped state, they

behave as an anisotropic, quasi-one-dimensional electronic structure with a moderate bandgap of 2–3 eV like a conventional semiconductor and they exhibit the electrical and optical behavior of semiconductors along with the mechanical action of typical polymers. When conjugated polymers undergo doping or photoexcitation, the p bond gets self-localized to undergo nonlinear excitation as polarons, solitons, bipolarons, etc., and the polymer transforms from a nonlinear excitation state to a metallic state [4]. Research on CPs is one of the hotspots in the field of materials due to their special polymeric structures and fascinating electrical, optical, mechanical, and biological properties, hence, they have been widely applied in many fields such as biotherapy, chemical/biological sensors, photo-/electrocatalysis, energy storage and conversion devices, electromagnetic shielding, and so on [5].

In comparison to conventional semiconductors, CPs possess unique bonding structures along the polymer backbones, consisting of alternating double (p) and single (s) bonds which endow them semiconducting properties. Once doped, the p-systems accommodate negative or positive charges that can pair with the dopants to form polarons, and can further be oxidized to bipolarons (supporting the transport of both holes and electrons) [6].

Conducting polymer nanocomposites are a class of hybrid materials synthesized using CPs with some inorganic materials of different sizes and nature using some chemical or electrochemical techniques giving rise to a new hybrid material with fascinating properties and significant application potential.

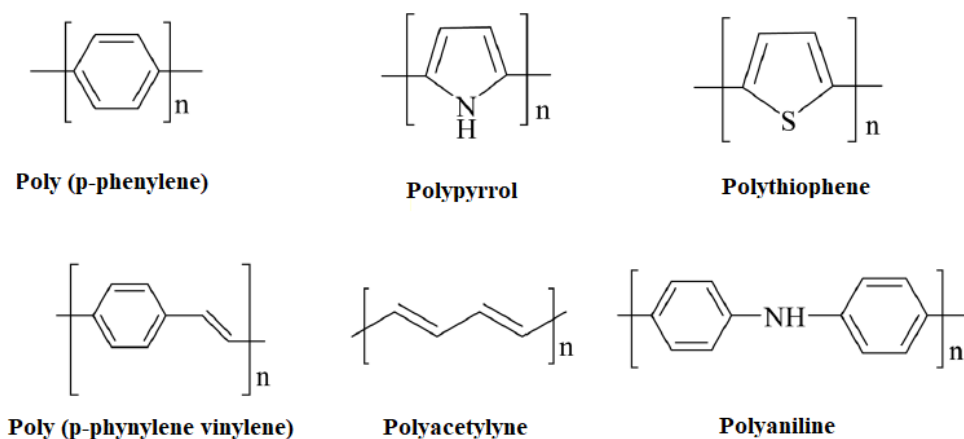


Figure 1. Structural illustration of different conducting polymers.

Conducting polymer nanocomposites has emerged as a boost and opened new doors for researchers in wide applications in the field of wastewater treatments, optoelectronic devices, batteries, electrochromic devices, biomedical materials, and devices such as sensing, imaging, etc [7].

Current research activities are highly focused towards the development of facile techniques for waste water remediation. Semiconductor based photocatalysis has been chosen as one of the promising methods for the degradation of toxic pollutants from the wastewater. Recently, CPs and its nanohybrids have attracted much attention due to their electrochemical properties, photo-stability, low cost, and efficacy. The nanohybrids of CPs with metal, metal oxides and other semiconductor materials have shown excellent photocatalytic activity for the degradation of toxic organic pollutants including dyes, pesticides and pharmaceuticals drugs [8]. In the present review, a brief overview of the concept of conducting polymer nanocomposites is provided with the synthesis methods, applications in various fields. Special emphasis has been given to application of

polyaniline-based nanocomposites in photocatalytic degradation of hazardous materials.

Conducting polymer nanocomposites

In comparison with conventional polymeric materials, conducting polymer nanocomposites (CPNCs) have gained increasing interests due to their improved electrochemical behavior, facile synthesis, biocompatibility, large surface area, high electrical conductivity, elevated carrier transport, and unique optical behaviors. Previously, multi-sided CPNCs with varying architecture and morphologies have undergone synthetization, to form nanomaterials including 0D (nanoparticles, nano-spheres), 1D (nano-ribbons, nanorods, nano-wires, nanofibers, nano-belts, and hollow structured nanotubes), 2D (nano-rings, nanosheets, nano-discs, nano-clips). Numerous physical and chemical techniques have been utilized for fabrication of nanocomposites, including physical template synthesis (i.e., hard and soft method), and template-free approach (i.e., self-assembly or interfacial polymerization, electrospinning, seeding approach) [9, 10].

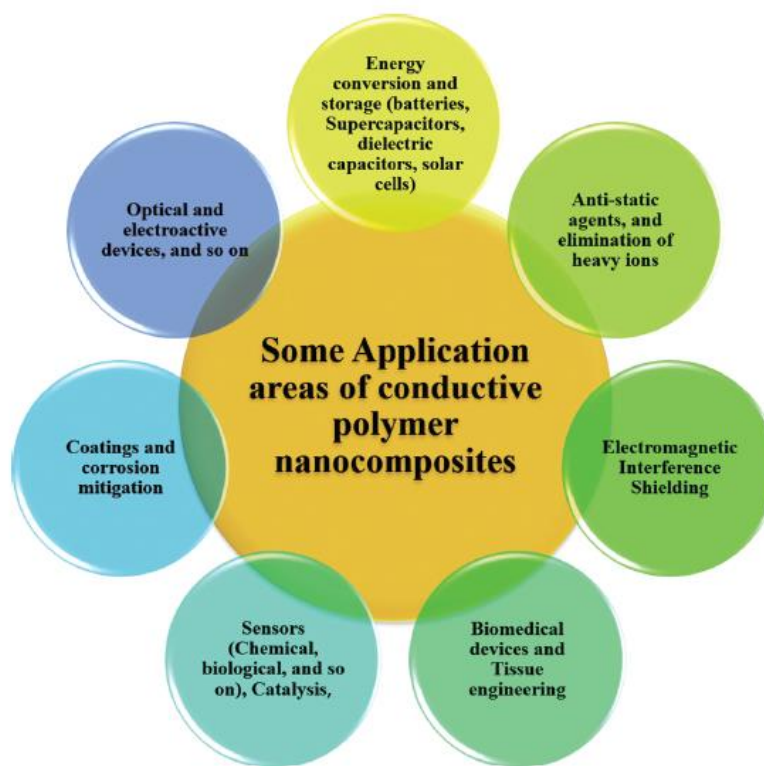


Figure 2. Some applications of CPNC [12]

Parameters to be considered in CPNCs development include charge transferring mechanism and charge mobility procedure during occurrence of redox reactions which result in excellent electrical behavior. Moreover, the doping procedure can notably escalate charge carrier entities, while enhancing conductivity. The basic attribute of CPs involves enlarged π conjugated mechanism along the polymeric chain which endows metallic propensity to their behaviors relative to electronics, magnetic and optical properties. Nevertheless, CPs also exhibit similar properties of conventional polymeric materials, such as good mechanical behavior, environmental stability, low temperature of processing, inexpensiveness, and so on. Moreover, it is essential to synthesize CPNCs to completely utilize inherent advantages in overcoming deficiencies, such as solubility. CPs ability to conduct and undergo solubility

is managed through modification of their chemical architectures [11].

Nanocomposites fabricated through CPs comingling with varying nanomaterials, including metallic entities such as Ag, Au and Pd, metallic oxides including Fe_2O_3 , MnO_2 , NiO , CuO , TiO_2 , WO_3 , ZnO , SnO_2 , V_2O_5 , RuO_2 , and chalcogenides including MoS_2 , WS_2 , Bi_2S_3 , CdS , CdSe , CdTe , along with carbon derivatives such as graphene (GN), graphene oxide (GO), carbon fibers (CF) and nanotubes (CNT), and other nil-conductive polymeric materials, have been broadly elucidated. Fabrication of CPNCs exhibiting elevated mechanical behavior, flexibility, and conductivities have demonstrated CPs capacity to serve as major material parts in light-emitting diodes, transistors, electrochromic devices, actuators, electrochemical capacitors, photovoltaic cells, and sensors. Enhanced properties of CPNCs have resulted broad applications in energy conversion and storages (batteries,

supercapacitors, dielectric capacitors, solar cells, fuel cells), chemical sensors, biosensors, catalysis, optical devices, electro-active devices, biomedical devices, electromagnetic interference shielding, corrosion protection, antistatic agent, removal of heavy metal ions, tissue engineering and other applications as schematically elucidated in Figure 2 [12].

Mechanism of CPs polymerization

Oxidative cross-linking of monomers is used in synthesizing CPs. From a polymerization perspective, the initial step is monomer oxidation resulting in formation of a radical cation, which subsequently reacts with another monomeric entity or radical cation, resulting in formation of a dimer. This is a vital approach in initiation procedure of polymerization. There are three main routes of initiation vis-a-viz, chemically initiated, electrochemically attained, and photo-induced oxidation, whereby individual routes showcase their own merits and demerits. From the perspective of the first approach, oxidation of the monomer is attained using chemically oxidizing entities such as ammonium persulfate or ferric chloride. From the perspective of the second approach, monomeric entities undergo electrochemical oxidization, while in third approach, light enables the monomeric oxidization using a photo-initiator [13].

Techniques used in synthesis of CP nanocomposites

Relative to mechanism and fabrication procedures, techniques utilized in fabrication of CPNCs may be categorized into three main classes: (i) ex-situ or sequestered synthesis, (ii) in-situ or sequential synthesis,

and (iii) single-pot or concurrent synthesis, as shown in Table 1 [14].

The major attribute of all the techniques detailed in the first classification is that the CPs and inorganic entities undergo individual synthetization, while hybridization is attained during next step via simplified or more intricate co-mingling of two or more entities where interfacial tension between varying entities determines notable properties of the resultant nanocomposite. These processes are simplified and normally highly appropriate for solution-oriented processing, which is a stepping-stone for enhancing mass production and processing using roll-to-roll printing. Added merit of this technique is the well-known synthesis step where each entity is individually fabricated using a well-established approach. Numerous other techniques exist for the hybridization procedure, ranging from very simplified approaches such as mechanical mingling of the two components, to highly intricate techniques such as ligand exchange preparation steps. Moreso, physical inculcation of CPs into inorganic nanomaterials can also be attained, resulting in highly aligned nanoarchitectures. However, this procedure is not simple because of the possibility of hindering the polymeric entities from penetrating the cavities of the nano-architecture host as a result of high interfacial tension because of physical constrains such as the hydro-dynamic polymeric radius, resulting in incomplete cavity filling where very poor pore filling ratios as low as 0.5% were attained [12, 15, 16].

Table 1. Techniques of synthesizing CP nanocomposites [12]

Main Group	Merit	Demerit	Example
Ex-situ synthesis	Simplified Solution processability	Restrained usage, inferior control of the contact between each component	Mechanical mixing Layer-by-layer deposition
In situ synthesis	Variable approaches relative to Chemical or electrochemical approach. Facile control of numerous parameters	Highly complex due to consideration of many parameters	In-situ polymerization. Electrodeposition. In situ reduction
Single pot synthesis	Simplified and brief processing duration	Constrained control over architecture and morphology of the products	Redox reaction Co-deposition

Properties and multifunctional applications of CP nanocomposites

CPNCs display enhanced properties such as improvement in batteries and super-capacitors electrolytic diffusion, enhancement in dielectric behavior of ferroelectric polymeric-oriented capacitors, facilitating efficient dissociation in solar

cells, minimization in response duration, and improvement in biosensors and chemical sensors sensitivity and increased corrosion inhibition. Additionally, the representative researches in CPNCs for specialized devices are also highlighted, as schematically illustrated in Figure 3 [17].

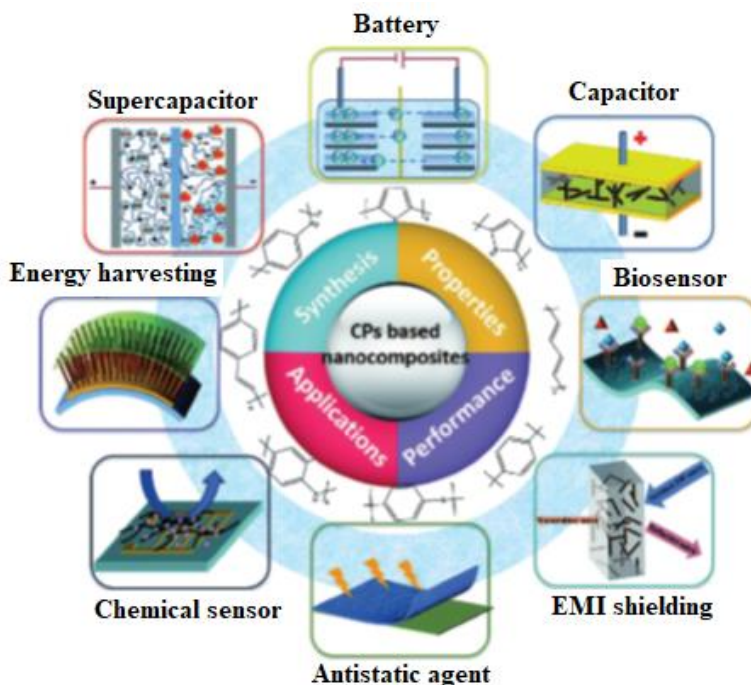


Figure 3. Elucidation of CPs nanocomposite applications [12]

Photocatalytic activity of CP nanocomposites

Water pollution has emerged as a serious problem all over the world due to the

increased agricultural, industrial and domestic activities. The quality of river and ground water has undergone immense deterioration due to the pollution by hazardous wastes. These toxic pollutants have significantly affected the human health and the quality of the ecosystem. The major sources of water pollution include heavy metal ions, dyes, phenols, polychlorinated biphenyls (PCBs), haloacetic acids (HAAs), pharmaceutical drugs, pesticides, disinfection byproducts (DBPs), and other synthetic chemicals [18].

Among different methods to removal of these pollutants, advanced oxidation process (AOP) has been extensively reviewed for complete decomposition of organic pollutants through the generation of reactive oxidants/radicals such as $\bullet\text{OH}$, $\text{O}_2^{\bullet-}$, etc. under different sources of light irradiation (UV, visible, microwave and solar light) [19]. Heterogeneous semiconductor photocatalysis is reported to be of low cost, eco-friendly as well as facile method for waste water treatment.

Inorganic metal oxides such as V_2O_5 , MnO_2 , WO_3 , Nb_2O_5 , SnO_2 , TiO_2 , ZnO , Fe_2O_3 , and Fe_3O_4 have been extensively reviewed in literature for water treatment processes. However, these oxides face some serious challenges in the large scale application process such as inactivity under visible light, faster electron-hole recombination and presence of a large band gap [20, 21]. Significant approaches have been implemented for band gap modification to prevent the recombination of photo-generated electron-hole pair and improve the photocatalytic under visible light. The utilization of CPs as sensitizing agents has been shown to rapidly improve the photocatalytic activity due to the presence of

p-conjugated systems containing electron rich species, which help in inserting electrons into the conduction band (CB) of most inorganic semiconductor oxides [22].

CPs possesses metallic conductivity and mechanical characteristics similar to metals/metal oxides. The conjugated one-dimensional structure of CPs offers outstanding electrical, optical and electronic properties, which are widely used for catalytic purpose [23]. Hence recent literature has been focused on the synthesis and application studies of the CP based nanocomposites, which show immense potential for environmental remediation in terms of facile synthesis, enhanced photocatalytic activity, reduced electron-hole recombination and variation of photocatalytic activity under UV-visible and solar light irradiation [24].

Photocatalytic mechanism of semiconductors under UV light irradiation

The photocatalytic activity of a material usually depends on its ability to create electron-hole pairs, which generate $\bullet\text{OH}$, $\text{O}_2^{\bullet-}$ that can attack the pollutants via secondary reaction. Photocatalytic reaction is usually subjected to the wavelength of light (photon) energy as the irradiation of light stimulates redox process of electron-hole generation [25]. Schematic representation of metal oxides-based semiconductors photocatalytic mechanism is shown in Figure 4.

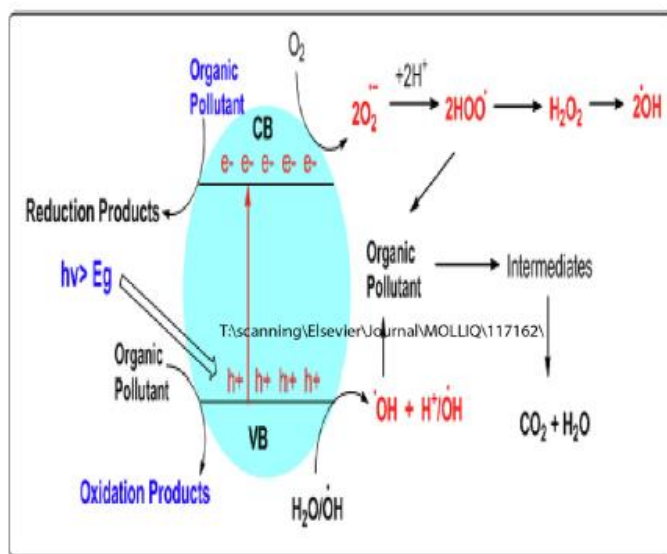
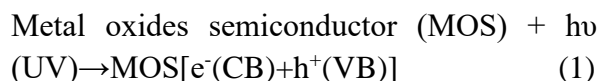


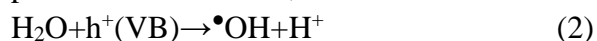
Figure 4. Schematic representation of metal oxides-based semiconductors photocatalytic mechanism [8].

The fundamental steps in the process of semiconductor photocatalysis are:

- Photoexcitation which is initiated by the photoelectrons that get promoted from the valence band (VB) of a semiconductor to the empty conduction band (CB) triggered by the source of irradiation.



- Ionization of water: This occurs when the holes at VB react with water molecules to produce $\bullet\text{OH}$ radicals, as shown below:

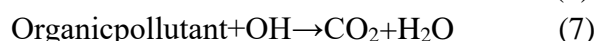


These radicals attack the organic pollutants and result in the formation of fragments.

- Oxygen ion sorption: in this step the electron (e^-) in the CB is taken up by the oxygen to produce $\text{O}_2^{\bullet-}$.



- Protonation of superoxide The $\text{O}_2^{\bullet-}$ radical undergoes protonation forming hydroperoxyl radical (HOO^\bullet) which degrades into hydroxyl radicals.



3.2. Role of conducting polymers (CPs) in photocatalysis

The rapid rate of photo-generated electron-hole pair recombination is prevented by the use of CPs as they help in tuning the band gap of semiconductors. As per density functional theory (DFT) calculations, CPs serve as low-band gap materials with wide adsorption spectra under visible light making them active photocatalysts under this source of irradiation [26]. Various metal oxides have been combined with CPs mainly comprising of PANI, PTH, PPy, poly(1-naphthylamine) (PNA), poly(o-phenylenediamine) (POPD), polycarbazole (PCz), polyacetylene (PA) etc. to improve their photocatalytic performance. CPs can act as p-type and form a p-n junction with metal oxide to control the electron-hole recombination [27]. The photocatalysis of contaminants with CPs and CP based nanocomposites exhibit the first-order reaction kinetics as reported in most of the studies.

Table 2. Photocatalytic characteristics of pristine conducting polymers [8].

CP	Contaminant (s)	Light	% Removal/ time
PNA	Methyl orange (MO)	Microwave	84%, 96%/ 15 min
PANI	Acid orange 7 (AO)	Microwave	65% /10 min
PEDOT	Metformin and PPCPs	UV light	99% /60 min
PNA nanotubes	Comassie Blue (CB) dye	Solar light	55% /60 min
PPy	Phenol	UV light	100% /4.5 h
Poly (3-hexylthiophene)(P3HT)	Phenol	UV and visible light	92% /240 min

Photocatalytic performance of pristine CPs

The studies conducted by the Riaz et al. [28], Ghosh et al.[29] and Yuan et al. [30] have shown that PNA, PANI, POPD, PCz, PEDOT and PPy nanostructures show excellent photocatalytic activity for removal of dyes and phenol. Kumar et al. [31] reported the photocatalytic degradation of mixture of PPCPs by PEDOT under UV light and found 99% degradation efficiency within very short time. Table 2 presents a comprehensive overview of photocatalytic activity of CPs.

Photocatalytic performance of CP based nanohybrids

The nanocomposites of CPs with metal oxides and other supporting material have shown excellent photocatalytic performance under different light irradiation. The hybrid material usually has high photo-response and delayed recombination of charge carriers. The hybrid system of CPs with metal oxides has distinct exciton-plasmon interactions and exhibit higher surface area for high absorption. Therefore, the current research focus has been on hybrid material development for increasing the efficiency of photocatalysis. PANI, PPy, PTh, PCz, PNA and PEDOT based nanocomposites are the most studied photocatalysts for waste water remediation.

Polyaniline based nanohybrids

PANI is one of most investigated conducting polymers among all CPs due to its high conductivity, facile synthesis procedure and good environmental stability. The research group of Riaz and coworkers [32] prepared PANI/SnO₂ nanohybrids via ultrasound assisted chemical polymerization using SnO₂ nanoparticles extracted from Psidium Guajava. The nanohybrids were tested as catalysts for the degradation of dichlorodiphenyltrichloroethane (DDT). Around 97wt% of DDT degradation was achieved within a very short period of 12 min under microwave irradiation. Similarly, Wang and Min [33], Chen et al. [34] and Reddy et al. [35] synthesized PANI/TiO₂ nanohybrids which showed higher photocatalytic efficiency as compared to pristine PANI or TiO₂ for the degradation of methylene blue (MB) as well as Rhodamine B (RhB) dye. The PANI/TiO₂ nanohybrids showed degradation efficiency of 81.74% for MB and 99 % for RhB under visible light irradiation. Gulce et al. [36] prepared PANI/CdO nanocomposites in aqueous solution of diethylene glycol via chemical oxidative polymerization method and investigated its photocatalytic activity against the MB and malachite green (MG) dyes under UV light and natural sunlight irradiation. Photocatalytic decolorization rate was increased up to 7 times by the addition of CdO with PANI. Around 99% degradation efficiency was achieved under natural

sunlight irradiation within 60 min. The nanocomposites of PANI with ZnO were also prepared by few researchers and these nanocomposites were reused multiple times in photocatalytic degradation process with a minimal loss of its catalytic reactivity [37]. Qin et al. [38] investigated the photocatalytic activity of MB dye under visible light irradiation. PANI/ZnO showed significantly enhanced photocatalytic properties, the reaction rate constant was found to be 0.01944 min^{-1} which was higher than the pure ZnO (0.00245 min^{-1}) and PANI (0.00585 min^{-1}). Mitra et al. [39] investigated the photocatalysis of dyes with aluminium-doped ZnO-PANI hybrids. They found that beyond an optimum weight percentage of Al-ZnO, there is a significant decrease in the photocatalytic activity of the hybrid. The synergistic effect of PANI and Al-ZnO led to higher photocatalytic activity. Cheng et al. [40] synthesized Ag-Cu₂O/PANI using a simple, low-cost process and results showed that the Ag-Cu₂O/ PANI exhibited a high long-term antibacterial activity against *S. aureus* and *P. aeruginosa*. Kadi et al. [41] developed core shell like CeO₂/PANI nanocomposites and the photocatalytic degradation efficiency of the synthesized nanocomposites was investigated against the degradation of nitrobenzene under visible light irradiation. The photocatalytic performance of CeO₂/PANI nanocomposite was found to be 6.6 times higher than PANI and 14.3 times higher than CeO₂. PANI based nanocomposites doped with SrTiO₃ were explored by Shahabuddin et al. [42] for the degradation of MB dye. Results showed that the degradation efficiency of nanocomposites which were doped with the SrTiO₃ nanocubes was higher than that of the undoped PANI and around 97% degradation

efficiency was achieved after 90 min under solar light irradiation. Shang et al. [43] and Wu et al. [44] explored PANI/bismuth oxide, PANI/Bismuth vanadate (BiVO₄), and PANI/bismuth niobite Bi₃NbO₇ nanocomposites. They noticed remarkable photocatalytic ability with high organic carbon content removal ability of the nanocomposite. Bu and Chen [45] investigated PANI/Ag/Silver phosphate (Ag₃PO₄) hetero-structure formed through in-situ deposition of Ag₃PO₄ nanoparticles on PANI chains and concluded that the nanocomposite had superior photocatalytic ability and stability because of an improved interfacial electric field resulting in profound electron-hole separation and migration efficiency. They found Ag formation on the PANI surface because of PANI assisted reduction of Ag⁺ to Ag⁰, which played an important role in the separation of photo-generated electron and holes. Studies on PANI/graphene and graphene oxides also reported excellent photocatalytic activity under visible light radiation for the degradation of toxic organic pollutants [46]. Zhang et al. [47] developed PANI nanorods arrays on graphitic carbon nitride (g-C₃N₄), and showed superior photocatalytic activity under visible light. Pandiselvi et al. [48] reported carbon nitride/polyaniline/ZnO composite with excellent photocatalytic ability under visible light for degradation of dyes.

Conclusion and future prospects

Conducting polymers and their nanocomposites have proven to be promising photocatalysts for decontaminating water. The role of CP based nanocomposites as photocatalysts are dependent upon the radical generation and transfer of photoelectrons to

CB of the metal oxides. Among pristine CPs, PANI was noticed to show high catalytic activity. Similarly, among PANI nanohybrids, superior catalytic activity was noticed for PANI/SnO₂. Hence the photocatalytic activity could be tuned to obtain optimum performance under UV/visible/solar or microwave irradiation. For future work, the design of metal ferrites/metal oxides nanoparticles via green routes and its nanohybrids with CP could be attempted and compared with the commercially available nanomaterials. The application of these green route derived nanohybrids materials is expected to add value to the knowledge pool of basic scientific research as well as technological development dealing with the designing of photocatalytic nanohybrids. These nanohybrids materials could also be developed at industrial scale for catalytic degradation of pollutants under microwave and visible light irradiation.

Acknowledgements

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