

Review

Evaluation of Polypyrrole as a Functional Sorbent for Water Treatment Technologies

Sylwia Golba *  and Justyna Jurek-Suliga

Institute of Materials Engineering, University of Silesia, 75 Pulku Piechoty Street 1A, 41-500 Chorzow, Poland; justyna.jurek-suliga@us.edu.pl

* Correspondence: sylwia.golba@us.edu.pl

Featured Application

The information provided in the review can be valuable and interesting for a broad audience of scientists involved in the synthesis of modern polypyrrole-based materials, as well as the groups that utilize it as a component of nanocomposites aimed at a sorbent functional unit. The knowledge of the relation between the synthetic path and morphological parameters opens the possibility to use it in reverse technology and tailor materials of the future with enhanced sustainability and efficiency of usage.

Abstract

Polypyrrole, which belongs to the conducting polymer family, has demonstrated profound potential in advanced water purification applications due to its inherent electrical conductivity, environmental stability, and tunable surface chemistry. As a sorbent, PPy exhibits high sorption capacity for aquatic contaminants, including heavy metals, pharmaceutical compounds, and their metabolites, as well as synthetic dyes. The removal efficiency is correlated to a complex interaction mechanism involving electrostatic attractions, redox activity, and π - π stacking. Recent advances have expanded the utility by further developing nanostructured PPy-based (nano)composites, which elevate sorption performance by increasing surface area, mechanical integrity, and selective affinity. In addition, its integration into membrane technologies has enabled the design of an effective filtration system with improved selectivity and regeneration capabilities. Moreover, PPy is effective in electrochemical processes of water treatment, including capacitive deionization and electrochemically assisted sorption, opening novel paths towards energy-efficient pollutant removal. The multifunctionality of PPy as a sorbent material highlights its value as an important material for water treatment, with the capability of extended modification tailored for emerging environmental needs revised in this work.

Keywords: polypyrrole; conductive polymeric material; sorption; specific surface area; water purification; drug removal; dye removal; metal removal



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1. Introduction

Polypyrrole (PPy) is a widely studied organic electroactive polymer with applications in conductive coatings, sensors, drug delivery systems, energy storage devices, and photothermal therapy for cancer treatment [1–3]. Its performance of PPy is strongly influenced by its electronic configuration and surface morphology, which can be tuned along with synthesis parameters [4]. Common synthesis methods include either chemical oxidative

polymerization, typically involving oxidants such as ferric chloride (FeCl_3) or electrochemical oxidation, in which the polymer is deposited under an applied anodic potential [5]. Additionally, unconventional techniques such as radiolytic polymerization, sonochemical synthesis, and enzyme-mediated processes have been proposed and explored [6]. The redox properties of PPy arise from both electron and mass transport phenomena, leading to structural transitions such as conformational rearrangements, swelling, shrinking, and relaxation under external stimuli [7]. Such dynamic properties make PPy a highly adaptable material for advanced functional systems. Moreover, as the polypyrrole becomes oxidized and acquires a net positive charge, counterions from the electrolyte are attracted electrostatically into the polymer matrix to maintain electroneutrality [8]. The balance is preserved either by the inward diffusion of cations or the outward expulsion of anions during redox cycling.

Polypyrrole exhibits optical and electrical properties comparable to semimetals, attributed to its delocalized π -electron system along the polymer backbone [9]. The electrical conductivity of the material is governed by the formation and mobility of charge carriers, known as polarons (radical cations) and bipolarons (dications), which facilitate charge transport through the matrix [10]. Also, doping plays an urgent role in modulating the electrical properties of PPy. Typically, small dopant anions, such as chloride ions, interact with approximately three to four pyrrole units, resulting in a dopant content constituting about 30–40% of the polymer's weight [11]. Various organic as well as polymeric dopants were employed, including dodecylbenzene sulfonate (DBS^-) [12], dodecyl sulfate (DS^-) [13], and polystyrene sulfonate (PSS^-) [14]. Both the nature and size of the dopant anion significantly influence the ion-exchange capability of PPy. Doping with small, mobile anions leads to a material of anion-exchange character, while doping with large, immobile anions transforms it into a cation-exchange material, as the immobilized anions within the polymer matrix necessitate the movement of cations to maintain charge neutrality [15]. Medium-sized anions provoke mixed ion-exchange behavior depending on the electrochemical environment.

The physicochemical properties of polypyrrole are highly dependent on the specific synthesis parameters employed, including the choice of synthetic method, substrate material, monomer concentration, additives, and dopants. These variables significantly influence the resultant PPy's morphology, degree of agglomeration, crystallinity, doping level, and charge storage capacity [16]. For instance, the use of different dopants has been associated with variations in the bipolaron-to-polaron ratio, which directly affects the conductivity of the material [11]. Moreover, the presence of surfactants or the application of sonication during polymerization has been demonstrated to influence the size, morphology, and electrical properties of the polymer. For example, the incorporation of sodium dodecyl sulfate (SDS) as a soft template during chemical polymerization affected the morphology and enhanced the electrical conductivity of the resulting polymer [17]. These findings underscore the importance of carefully controlling synthesis parameters to tailor the properties of PPy for specific applications to meet the requirements of technological applications.

Scientific interest in polypyrrole is driven by its physicochemical properties, including high environmental stability, redox activity, ease of synthesis, and tunable surface functionality [18]. These features make the polymer a choice for a wide range of sorption-based applications in environmental remediation, sensing, and separation technologies. The polymer's sorption efficiency is associated with its structural characteristics, such as surface area, porosity, and doping level, which can be modulated at the synthesis stage and in post-treatment strategies. Furthermore, its ability to interact with ionic, polar, and non-polar species through electrostatic interactions, hydrogen bonding, π - π stacking, and redox-mediated mechanisms contributes to its utility as a sorbent material. This review

aims to provide an examination of the sorption behavior of polypyrrole, with a focus on the underlying mechanisms of adsorption and ion exchange, and its practical implementation in fields such as water purification and pollutant removal. The review explores the role of structural optimization, including controlled porosity, tailored doping, and hierarchical architectures, in facilitating rapid ion diffusion and improving regeneration performance. Special emphasis is placed on recent advancements, highlighting novel synthesis techniques, functionalization approaches, and the development of polypyrrole-based composite materials that enhance sorptive performance along with selectivity.

2. Sorbent Synthesis

The synthesis of polypyrrole (PPy) has been extensively reviewed in the scientific literature, with particular emphasis on the influence of variations in synthetic approaches on the resulting morphology, structural features, and functional performance of the polymers. Numerous recent reviews have systematically explored the impact of synthesis parameters—including the choice of oxidizing agents, polymerization method (chemical vs. electrochemical), dopant type, reaction time, and temperature—on the physicochemical properties of PPy, such as conductivity, porosity, surface area, and electrochemical activity [16,19]. These studies demonstrate that fine-tuning synthesis conditions can yield polypyrrole materials with diverse architectures, ranging from nanofibers and nanoparticles to porous films and aerogels, suited to specific applications such as energy storage, sensors, and environmental remediation [20]. In particular, electrochemical polymerization offers precise control over film thickness and surface morphology, making it favorable for applications requiring well-defined interfaces [21]. Nanofibers of the polypyrrole were positively verified as supercapacitors [22], sensors [23], electromagnetic shielding [24], tissue engineering, neural regeneration, and stimulation [25]. The superior performance can be attributed to the large specific surface area, network architecture, elevated electrochemical activity, tunable bandgap, and inherent biocompatibility [18]. Several techniques were utilized for polypyrrole nanofiber production, including electrospinning [26], interfacial polymerization [27], soft templating (Figure 1) [18], and seeding [28]. In the production process, several output parameters need to be managed, including specific surface area, both morphological and structural parameters like electronic structure, length, diameter, flexibility, and conductivity of the nanofibers.

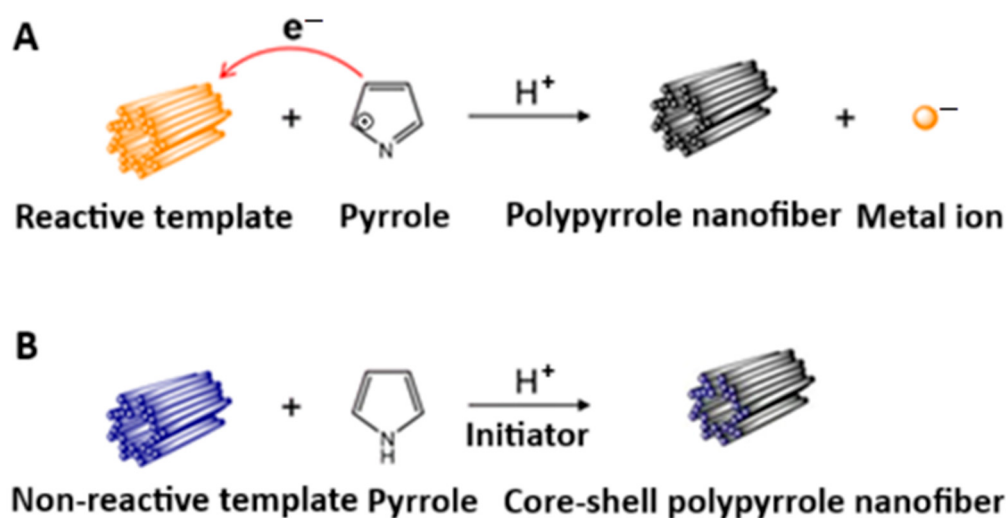


Figure 1. Illustration for the synthesis process of PPy nanofibers using (A) a reactive template and (B) a non-reactive template (reproduced from [18] under the terms and conditions of a Creative Commons Attribution-Non Commercial 3.0 Unported Licence).

The nanofibers can assemble in 1D, 2D, and 3D microstructures that serve to produce various functional forms, including microfibers, sheets, and membranes. Reviews highlighted the role of surfactants and templates in directing nanostructure formation, enabling the fabrication of hierarchical or porous PPy structures with enhanced functional performance [29]. These analyses provide insights into the structure–property relationships of PPy, guiding the rational design of polymeric systems for targeted technological applications.

The performance of polypyrrole as a sorbent material is influenced by the synthesis route and associated processing parameters, which govern the physicochemical characteristics. Parameters such as electrical conductivity, surface morphology, porosity, and crystallinity are particularly sensitive to variations in polymerization technique, monomer and dopant concentrations, temperature, and the presence of surfactants or templating agents [30]. Conductivity enhanced by doping (oxidative or electrochemical one) impacts the redox activity and ion-exchange capacity of PPy, essential for the sorption of charged species [31]. Surface morphology and porosity, which determine the accessible surface area for adsorption, can be tuned by adjusting the polymerization rate, reaction medium, or introducing soft/hard templates [32]. Furthermore, the degree of crystallinity influences the mechanical stability and diffusion characteristics within the polymer matrix, thereby affecting the kinetics and reversibility of sorption processes [33]. Hence, the optimization of synthetic parameters is necessary to tailor PPy-based materials aimed at specific sorption applications.

3. Application Areas

Polypyrrole emerged as a versatile sorbent material owing to its redox activity, environmental stability, and ability to interact with a wide range of analytes through multiple interactions, including electrostatic, π – π , or hydrogen bonding. The high affinity for both organic and inorganic contaminants made it useful in water treatment, environmental remediation, and sensor development. The sorption capabilities of PPy are enhanced by tuning its porosity and surface area, which increases the accessibility of active sites for target molecules [34]. Similarly, ultralight 3D aerogels formed with PPy on cellulose acetate nanofibers showed a large specific surface area and high porosity that led to an adsorption capacity of Cr(VI) equal to 244.65 mg/g and enhanced mechanical properties (compressive strength of 14.49 kPa) [35]. Nanocomposites based on PPy enriched with activated carbon or graphene oxide exhibited synergistic effects, taking advantage of both the conductivity and versatility of the polymer and high surface area and microporosity of the carbon component [20,36]. These findings underscore the importance of porosity engineering in optimizing the performance of novel sorbent materials. The following paragraphs focus on the urgent application fields, including water purification in terms of metal, drug, and dye elimination, followed by bacteria removal and membrane separation aspects.

3.1. Water Purification

Water contamination represents a primary contributing factor to a wide range of emerging human health concerns. Substances like dyes [37], heavy metals [38], and antibiotic drug metabolites [39] are excreted daily, becoming the major source of pollution in the environment. The presence of impurities in an aqueous environment affects its quality due to strong environmental persistence, biological activity, and bioaccumulation. These lead to deterioration of water quality determined by the parameters like biochemical or chemical oxygen demand (BOD or COD, respectively), emissivity, salt concentration, and photosynthetic capacity [40].

There are several methods for the removal of impurities from wastewater using biological, chemical, and physical approaches. Adsorption, ion exchange, membrane diffusion,

coagulation, flocculation, and precipitation belong to physical methods. Irradiation and the Fenton oxidation process correspond to chemical ones. Microbial decomposition and degradation using agricultural biomass involve biological processes. For all methods, one can point out disadvantages, such as issues associated with chemical or biological residue, issues associated with membrane utilization, including fouling and clogging, as well as the potential toxic impacts of contaminants on biological systems [41,42]. Some of these methods are highly effective; however, their usage requires high operating costs and may produce toxic by-products. In this aspect, the adsorption process is associated with high cost efficiency, environmental friendliness, and versatility in removing toxic chemicals from wastewater. Additionally, the method does not need complicated equipment, and as the reaction kinetics are high, it brings validated capacity to remove a wide range of pollutants [43,44]. Being objective, one should point out some challenges in the adsorption technology, like the fabrication of novel adsorbents featuring high adsorption capacity and stability [45]. Also, the recycling of adsorbent and its reusability are crucial in terms of commercial-scale usability.

A key determinant of the adsorption process is the selection of an efficient adsorbent. The adsorption capacity of an adsorbate is governed by multiple parameters, including the surface area of the adsorbent, the nature of adsorbate–adsorbent interactions, the mass ratio and particle size of the adsorbent, as well as physicochemical conditions such as temperature and pH [46]. The preferred characteristics, including high surface area, elevated porosity, and substantial adsorption capacity, should be complemented by mechanical robustness, economic feasibility, and environmental sustainability. Some of the most extensively used adsorbents are activated carbon and silica gel [47] owing to their substantial surface area and advantageous adsorption characteristics; nevertheless, their elevated cost and challenges associated with regeneration have prompted the scientific community to pursue alternative solutions.

In terms of the kinetics of the adsorption process, several models describing the experimental data are available, like the pseudo-first-order, pseudo-second-order, and film diffusion and intraparticle diffusion ones [48,49]. Several parameters directly influence the extraction efficiency, concerning both sample (sorbent quantity, sample pH) and process character (solvent desorption, desorption time, ionic strength, or contact time) [50,51]; hence, they have to be investigated and optimized for a particular material. The pH value of the aqueous solution affects the surface charge of the adsorbent, the degree of ionization, and the selection of adsorbate during adsorption. The efficiency of ion adsorption might also be influenced by interfering ions; hence, such competitive reactions shall be taken into consideration [52].

An emerging field focuses on the application of nanoadsorbent materials, which attract growing interest because their size and unique physicochemical properties offer numerous advantages [53]. In this area, one group of adsorbent materials comprises nano-sized metal oxides, including Al_2O_3 , Fe_3O_4 , and MgO . Their efficiency is dictated by traits like superior surface reactivity, dictated by high surface area and profound adsorption capacity in comparison to the commercial analogues [54,55]. More effective contaminants extraction is commonly noted under extreme values of pH [56]; hence, the composition of sorbents is modified to shift the solution pH to the neutral range, e.g., for composites composed of metal oxide with conducting polymer [57]. In the field of drug removal, various materials were tested as adsorbents, including powder of activated carbon, active carbons prepared from bamboo, carbon nanotubes, canola, chitosan, CNTs, fly ash, chitin, zeolite, peanut hull, and magnetic Fe_3O_4 /chitosan nanoparticles, additionally mesoporous adsorbents including silicates and Al_2O_3 [58–60]. However, one must also be aware that prolonged exposure to nanoadsorbents might create environmental challenges, and their recycling remains difficult because nanoadsorbents do not separate easily from aqueous solutions through filtration or centrifugation [61].

Conducting polymers exhibit chemisorption when a chemical species is formed through electronic interaction between the adsorbent and the adsorbate [62]. CPs, including PPy, might be used as powder or surface coating dedicated to extraction applications [63]. Sorbent characteristics dictated by traits like hydrophobicity, π -interaction capability, acid-base behavior, the presence or absence of polar functional groups, ion-exchange properties, hydrogen-bonding ability, environmental stability, and ease of synthesis directly determine the efficiency of separation [64]. Being realistic, one shall point out some drawbacks of CPs like low mechanical properties [65], relatively poor thermal stability in air [66], and non-biodegradable nature [67] that restrict the practical applications. For most pure PPy samples, low adsorption capacity is induced by the agglomeration provoked by π - π bond formation [68]. To overcome this obstacle, novel composites were proposed with components like metal oxides or metal ferrites, e.g., magnetic CNTs-CoFe₂O₄ [34,69] for methylene blue removal. Also, the addition of mineral mesoporous materials [70] elevated the adsorption capacity.

3.2. Sorption of Metal Ions

Rapid industrialization and population growth generate substantial volumes of wastewater containing heavy metal ions that degrade the aquatic environment [71]. Heavy metals are the cause of a significant environmental threat because they exhibit high toxicity and accumulate and biomagnify within biological systems [72]. Another consequence of heavy metal presence in the environment is their role in the disruption of the natural food chain via bioaccumulation [73,74]. Inorganic pollutants are characterized by traits like solubility, oxidation-reduction potential, and complex formation. Being discharged into water, they are converted to hydrated ions that disrupt the enzymatic process with enhanced absorption into the cell organism [75]. Methods like precipitation, electrochemical treatment, photocatalytic reduction, and adsorption have been utilized to reduce Cr(VI) [76]. Still, an adsorption method offers the most promising approach because it uses inexpensive, sustainable, and renewable materials as adsorbents [77].

Various sorbents were verified in terms of sorption ability for heavy ion removal. These included magnetic starch-g-poly(acryl amide)/graphene oxide nanocomposite [78], poly(n-vinylpyrrolidone) modified clay [79], genipin cross-linked chitosan/poly(ethylene glycol), activated carbon prepared from date stones [80], carbon nitride [81], composites of graphene and layered double hydroxides [71], g-C₃N₄-based materials [82], geopolymers [83], chitosan [84], and wheat bran [85]. Metal removal from wastewater was also realized with the application of various adsorbents of ions obtained by immobilizing functional groups on the surface of the substrate, including mesoporous silica [86], activated carbon [87], and polymers [76]. Both mesoporous silica and activated carbon have been conveniently applied as adsorbents owing to the very large surface areas, commonly greater than 1000 m²/g. Still, the manufacturing processes for such sorbents are complex and expensive. Another group of promising sorbents is composed of coordination polymers [88] that contain nitrogen, oxygen, or sulfur atoms as binding sites. Chemical stability and good ion exchange performance make PPy suitable for the removal of Cr(VI) [89], which is one of the most toxic contaminants in water [90]. PPy's usefulness in the area of metal extraction comes from its nanostructure [91]. During the synthesis, polypyrrole tends to agglomerate into particles with a specific surface area of 12.21 m²/g and the adsorption capacity of Cr(VI) of 21.87 mg/g [92]. Details of the synthesis for the described sorbents, along with their morphological and some physical properties, are depicted in Table 1. Polypyrrole-based adsorbent for the removal of arsenic ions in a batch equilibrium system was proposed by Hqu [93]. PPy powder acted as an effective sorbent for the removal of arsenic ions at the optimum conditions of pH (6.5) and a contact time (6 h). Metallic co-ions, namely, zinc and cadmium, had a negligible effect on the adsorption efficiency

under the optimum conditions; hence, the adsorbent displayed preferential adsorption of As. It was pointed out that the presence of amine groups in PPy played the key role in the adsorption of arsenic ions [93]. Dodecylbenzene sulfonate (DBS) doped polypyrrole was used to remove radioactive cesium from aqueous solution [94]. Among radionuclides, radioactive ^{137}Cs represents a major component of nuclear waste and draws particular interest because it emits intense gamma rays and possesses a long half-life of 30.17 years, making its exposure highly harmful to humans [95]. The adsorbent rapidly and efficiently adsorbed the ^{137}Cs radionuclide, achieving a maximum sorption capacity of 26.2 mg/g at 40 °C. The thermodynamic analysis showed an endothermic and spontaneous nature of the adsorption process. Applied gamma irradiation showed no significant effect on the adsorption performance, even with the gamma radiation dose of 200 kGy [94].

Although the fabrication process of these materials is not complex, their relatively low surface area leads to inefficient extraction of heavy metals from wastewater [88]. The alternative candidates for the role of efficient heavy metals sorbent are nanocomposites with enhanced, more developed surfaces. They can be classified into different classes, including PPy-metal oxide hybrids, PPy-carbon-based nanomaterials, and bio-template-assisted systems. In the PPy-metal oxide hybrids group, colloidal nanocomposites of PPy/hollow mesoporous silica (PPy/HMSNs) were characterized with enhanced adsorption of Cr(VI) ions from aqueous solutions [96]. Also, a PPy/silica nanocomposite obtained via templated synthesis was used as a sorbent for heavy metal ions (Hg^{2+} , Ag^{+} , and Pb^{2+}) [97]. A bottom-up approach produced samples with a cratered surface, and using 4 mL of silica sol solution (Ludox SM-30) yielded the nanocomposite with the highest BET surface area of 306 m^2/g . As determined using the BET method, the amount of loaded silica solution controls the surface areas of the PPy/silica nanocomposites, as it increased over three times after the addition of the silica component. The PPy/silica nanocomposite prepared with SM-30 silica solution (4 mL) exhibits a type IV-like N_2 adsorption–desorption isotherm with hysteresis as typically shown by mesoporous-type materials [98]. The Barrett–Joyner–Halenda method, applied to the adsorption branch of the N_2 isotherm, determined the pore size distribution with the same pore size as the PPy/silica nanocomposite, with an average silica particle size of 7 nm, as in the original silica [97]. The adsorption capacities of the PPy/silica nanocomposite indicated that the material exhibited a higher complexation affinity for several metal ions (termed as “soft”) (0.97 (Hg^{2+}), 0.75 (Ag^{+}), and 0.53 (Pb^{2+}) mmol/g) than the “hard” metal ions (0.02 (Cu^{2+}), 0.03 (Ni^{2+}), 0.01 (Cd^{2+}), and 0.01 (Cr^{3+}) mmol/g) (Figure 2). The complexation reaction between specific ligands and target ions [99,100] governed the selectivity, as soft acids such as Hg^{2+} readily form stable complexes with soft bases like the secondary amines in pyrroles [97].

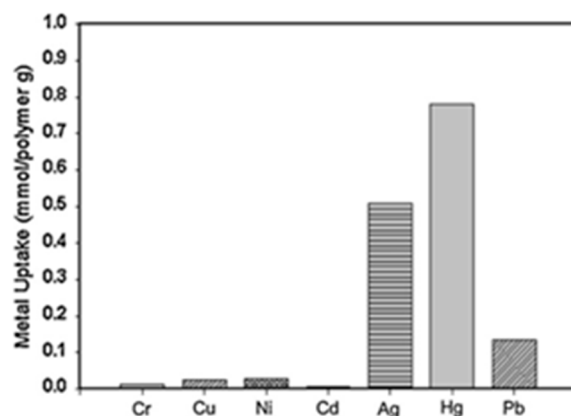


Figure 2. Metal ion adsorption capacities for PPy/silica nanocomposites (SM-30, 4 mL) (reproduced with permission from [97]; published by Elsevier, 2012).

An approach to extend the PPy's specific surface area, and hence to improve the adsorption performance, is template usage. Fang et al. [101] synthesized graphene/SiO₂@PPy nanocomposites with a specific surface area of 37.6 m²/g and an adsorption performance of Cr(VI) equal to 429.2 mg/g. The removal mechanism relied on electrostatic attraction, ion exchange, and the reduction process of partially adsorbed Cr(VI) to Cr(III) [101]. A nanocomposite of PPy/SBA-15 was used for Hg(II) adsorption from aqueous medium [102]. Mesoporous SBA-15 silicate was an adsorbent due to its high surface area, high pore volume, and ordered structure to functionalize the surface [103]. Optimum values were found as pH 8, contact time of 60 min, and the amount of adsorbent of 1 g/L. The results showed agreement with Langmuir adsorption and high adsorption capacity (200 mg/g). The N₂ adsorption/desorption isotherms of materials (Figure 3) revealed that the SBA-15 is a mesoporous material, as the isotherm of type IV was recorded. Such behavior was pictured with a hysteresis loop of type H1 and a steep increase in adsorbed N₂ at relative pressure $p/p_0 = 0.6$ – 0.8 [104]. The BJH pore size distribution curve revealed a narrow pore size distribution with an average pore size of 8.2 nm for SBA-15. The specific BET surface area of SBA-15 was 753.5 m²/g, while it decreased to 97.6 m²/g for PPy/SBA-15. For the nanocomposite, the inflection point of the isotherm shifted to a lower p/p_0 value, which supported the PPy insertion into the channels of SBA-15 [102].

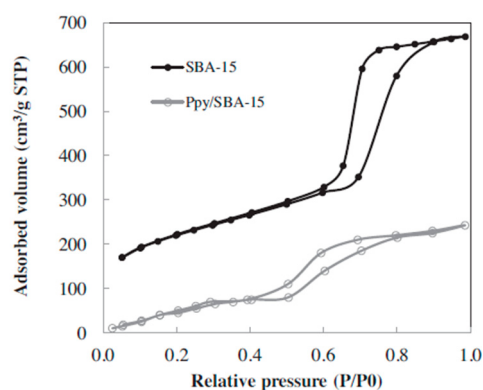


Figure 3. N₂ adsorption/desorption isotherms of SBA-15 and PPy/SBA-15 (Reproduced with permission from [102]; published by Elsevier, 2016).

Sodium dodecylhydrogensulfonate, poly(vinyl pyrrolidone), and poly(vinyl alcohol) were tested as surfactants in PPy synthesis aimed at the preparation of a sorbent for Cd removal [105]. Additionally, nanocrystalline Al₂O₃ was dispersed in the matrix. The sorption characteristic of nanocomposites was verified in the batch method. It was the Langmuir model that data were better fitted to, while increasing the pH above 5 substantially reduced the amount of sorbed Cd(II), while using PVP as a surfactant achieved the maximum Cd(II) removal across various concentrations [105].

The PPy-carbon-based nanomaterials group includes materials served by Ko et al. [106], who prepared PPy/carbon black composite particles. The specific surface area of 64.47 m²/g, and the adsorption capacity reached 174.8 mg/g. Pyrrolic nitrogen induced reduction in hexavalent chromium to trivalent chromium, while trivalent chromium adsorbed to oxidized pyrrolic N through covalent bonding interactions. Hexavalent chromium adsorbed to pyrrolic N through hydrogen bonding interactions [106]. In a search for an efficient adsorbent, nanofibers were used as templates to further improve the accessibility of adsorption sites. PPy-wrapped oxidized MWCNTs nanocomposites and bamboo-like PPy nanotubes exhibited adsorption capacities at 294.1 mg/g [107] and 556.8 mg/g (9.28 mmol/g) [108], respectively. A cobalt oxide-graphene nanocomposite functionalized with PPy with a heterogeneous structure was tested as a heavy metal, namely Pb(II)

and Cd(II) sorbent [75]. A heterogeneous porous structure with a specific surface area of $133 \text{ m}^2/\text{g}$ enhanced the adsorption process. The optimal pH of sorption, recognized as a highly efficient process, was found as 5.5 (93.08%) for Pb(II) and 6.1 (95.28%) for Cd(II), respectively. The maximum adsorption capacity (Q_{max}) reached 780.3 mg/g and 794.2 mg/g for Pb(II) and Cd(II) for spontaneous and endothermic processes. Hysteresis loops recorded in the isotherms characterized as type IV manifested the presence of mesopores in the materials. A higher number of adsorption sites, pore sizes, and volume enhanced the diffusion and supported the adsorption capacity of the pollutants for the nanocomposite. The adsorption ability increased with the rise in pH to the optimal value and then decreased, which reflected the interplay between sorbent components and H^+ and OH^- ions governed by electrostatic attractions and the coordination of lone pairs of nitrogen and oxygen atoms (Figure 4) [75].

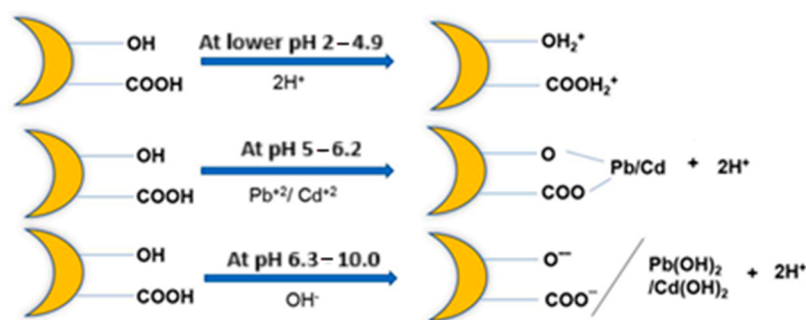


Figure 4. Proposed mechanism of the metal sorption: Pb(II) and Cd(II) (reproduced with permission from [75]; published by Elsevier, 2021).

The bio-template-assisted systems are represented by nanofibrillar composites composed of polypyrrole/bacterial cellulose (PPy/BC) tested for hexavalent chromium (Cr(VI)) removal [109]. A 3D nanofiber network structure of BC [110] provided an increase in the strength and stability of the material, which made it useful as a chemical reaction template [111]. Multiple OH groups of BC were utilized to form hydrogen bonds with pyrroles' -NH- groups, as a strategy to prevent the agglomeration of PPy particles aimed at improving the adsorption capacity. A core-shell structure was formed when PPy was wrapped on cellulose fibers. The composite removed Cr(VI) efficiently with an adsorption capacity of 555.6 mg/g at optimal conditions, with adsorption data conforming to the Langmuir isotherm model and the pseudo-second-order model. The act of Cr(VI) adsorption on the composite was provoked by ion exchange and electrostatic interactions. It was also confirmed that pyrrolic nitrogen reduced a portion of Cr(VI) to trivalent chromium (Cr(III)) and retained it on the composite surfaces through chelation [109]. The composite's performance as an adsorbent can be tuned by reducing the cellulose size to the nanoscale, expanding its specific surface area, and shortening the intraparticle diffusion distance. In such a system, PPy enhances the mechanical strength and stability of the system [112].

The adsorption and Cr(VI) detoxification capability of polypyrrole-bacterial extracellular polysaccharide (PPy-EPS) nanocomposite was tested by Kamala-Kannan [113]. Bacteria secrete extracellular polymeric substances (EPSs) during growth, forming biopolymers composed of proteins, lipids, glycoproteins, and sugars [114]. As bacterial EPS was reported to exhibit high adsorption potential for dyes and heavy metals [115,116]; hence, the nanocomposite with EPS was prepared in chemical oxidative polymerization. The surface area of the PPy-EPS was $26.21 \text{ m}^2/\text{g}$, and it reduced 80% of Cr(VI) within 30 min [113]. A high reduction rate was reported for the reduction of Cr(VI) into Cr(III) (Figure 5), explained by the high surface area that enhanced the interaction between Cr(VI) and nanocomposite,

coupled with the action of biomolecules' functional groups supporting the Cr(VI) proneness to interact [113].

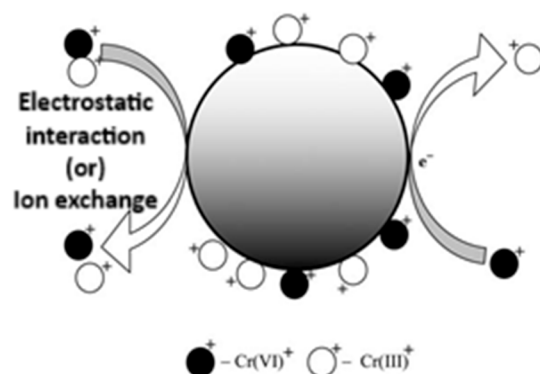


Figure 5. Schematic representation of Cr adsorption and Cr(VI) reduction onto PPy-EPS nanocomposite (reproduced with permission from [113]; published by Wiley, 2016).

Also, glycine-doped polypyrrole (PPy-gly) adsorbent was used for the removal of Cr(VI) [114]. The adsorption process was pH dependent, with a higher removal efficiency of PPy-gly than PPy homopolymer. The isotherm data closely followed the Langmuir isotherm model, showing a maximum adsorption capacity ranging from 217.39 to 232.55 mg/g. The adsorption process was dictated by the electrostatic interaction provoked by glycine's protonated amine groups and HCrO_4^- anions at low enough pH [114].

Another approach for the effective removal of hexavalent chromium Cr(VI) from water was provided by Alsaiani [115] through the fabrication of magnetite-based polymeric nanocomposite composed of magnetic Fe_3O_4 nanoparticles immersed in nitrogen-rich polymer matrix including chitosan (CS) and polypyrrole (PPy). The kinetic study revealed a good fit of the adsorption data with the Langmuir isotherm, interpreted as monolayer adsorption of Cr(VI), while the maximum adsorption capacity of Cr(VI) was 105 mg/g. The removal mechanism is based on chemical reduction partnered with adsorption. The co-existing ions effect showed that sulfate was the most affected co-ion, while bicarbonate participated to a smaller extent [115]. Nitrogen-type chelating resins appear promising owing to their high adsorption property for heavy-metal ions. Hence, the nitrogen atoms embedded in macrochains were used as active adsorption sites for nickel ion adsorption when polyethyleneimine with polypyrrole was produced [116]. The BET surface area of 17 m^2/g was reported along with the adsorption isotherm curve of type II. The average particle size of the nanocomposites of 18–34 nm was measured using the FESEM micrograph. The Ni ion adsorption efficiency increased markedly from 42% for pristine polypyrrole to 99.8% for PPy-PEI nanocomposite in a batch equilibrium system [116]. The rose leaf modified by coating with PPy served to remove Pb^{2+} and Cd^{2+} from aqueous media [117]. The plot of kinetic data of the ions' adsorption followed the pseudo-second-order kinetic model, while the adsorption data were fitted with the Freundlich model. The metal removal percentage was related to adsorption conditions, with the maximum efficiencies of 81% and 92% observed for Pb^{2+} and Cd^{2+} at optimum conditions [117]. A polypyrrole/corn cob bioreactor immobilized with *Zoogloea* sp. was designed for the degradation of several entities, including 17 β -estradiol (E2), nitrate, and Mn(II) ions [118]. It used the microorganism immobilized (MI) technique that combined the advantages of microbial activity and material characteristics [119]. Immobilizing microorganisms on a carrier in the MI bioaugmentation method increases cell density, enhances bioactivity, and improves resistance to complex contaminated groundwater environments compared to using free cells. In the study, the removal efficiencies of E2, nitrate, and Mn(II) at the optimal operating conditions were calculated as 84.21%, 82.96%, and 47.91%, respectively. The removal efficiencies were

dictated by the formation and action of biomanganese oxides, while biological precipitation's analysis showed a dominant role of adsorption and redox conversion on the oxide surface for the impurity removal [118]. PPy exhibits considerable potential as an advanced sorbent material for metal ion removal when compared to conventional sorbents such as activated carbon, zeolites, silica gel, and metal oxides. The traditional sorbents offer high surface areas and moderate sorption capacities; still, they also often lack specificity, exhibit poor performance in complex matrices, and require energy-intensive or chemically harsh regeneration processes. In contrast, PPy offers advantages stemming from its inherent electroactivity, tunable surface chemistry, and redox-responsive behavior. The redox-active nature of PPy enables reversible modulation of its charge density, allowing for electrochemically controlled sorption and desorption of metal ions without the need for external reagents. This attribute facilitates efficient and sustainable regeneration, particularly favorable for repeated use in water treatment systems. Furthermore, PPy demonstrates high selectivity toward specific metal ions even in complex aqueous environments containing competing species, due to its ability to form coordination complexes and its adaptable doping strategies.

Table 1. Comparison of the synthetic and physicochemical details of PPy-based sorbents aimed at metallic ions removal.

Sorbed Metal	Description	Dopant	Morphology	S BET [m ² /g]	Adsorption Capacity [mg/g]	Optimal pH	Adsorption Isotherm	Ref.
Cr(VI)	3D aerogels with PPy on cellulose acetate nanofibers	SO ₄ ^{2−}	porous structure with cross-linked nanofibers		244.65 31.92	2.0	Langmuir	[35]
Cr(VI)	sonochemically synthesized	Cl [−]	a fused globular	12.21	21.87			[92]
As(V)	black PPy powder	Cl [−]	cauliflower-like porous	10.27	1.91	6.5	Freundlich	[93]
137Cs	black PPy powder	DBS [−]	globular particles	11.28	26.2	5.0	Langmuir	[94]
Cr(VI)	colloidal nanocomposites of PPy/hollow mesoporous silica	Cl [−]	PPy wrapped on the hollow mesoporous silica	325	322	2.0	Langmuir	[96]
Hg ²⁺ , Ag ⁺ , and Pb ²⁺	PPy/silica nanocomposite	Cl [−]	cratered surface	85 (no silica) 306 (46 wt% of silica)	0.97 (Hg ²⁺), 0.75 (Ag ⁺), and 0.53 (Pb ²⁺) mmol/g			[97]
Cr(VI)	graphene/SiO ₂ @PPy nanocomposites	SO ₄ ^{2−}	porous film made with nanospheres	17.6 (PPy) 37.6 (graphene-silica-PPy nanocomposite)	429.2	2.0	Langmuir	[101]
Hg(II)	PPy/SBA-15	Cl [−]	PPy inside and outside of the SBA-15 pores	97.6	200.0	8	Langmuir	[102]
Cd	PPy synthesized in the presence of surfactants (PVP, PVA)	Cl [−]	globular structure, with the size of particles dependent on the type/concentration of surfactant			5	Langmuir	[105]
Cr(VI)	PPy/carbon black	SO ₄ ^{2−}	core-shell structure	64.47	174.8	3		[106]
Cr(VI)	PPy/oxidized MWCNTs nanocomposite	Cl [−]	PPy deposit on the surface of the OMWCNT	6.1 (PPy) 201.4 (OMWCNT) 34.1 (PPy/OMWCNTs NCs)	249.18	2	Langmuir	[107]
Cr(VI)	PPy with electrospun V2O5 nanofibers as templates	Cl [−]	rough surface of PPy nanotubes with the outer diameter 60–280 nm		482.6 9.281 mmol/g		Langmuir	[108]
Pb(II), Cd(II)	cobalt oxide graphene PPy nanocomposite	Cl [−]	oxide uniformly surrounded by PPy supported by GO	133	780.3 for Pb(II) 794.2 for Cd(II)	5.5 for Pb(II) 6.1 for Cd(II)	Langmuir and Temkin	[75]
Cr(VI)	PPy/bacterial cellulose (PPy/BC)		core-shell of PPy wrapped on cellulose	95.9	555.6	2		[109]
Cr(VI)	PPy-bacterial extracellular polysaccharide (PPy-EPS) nanocomposite	Cl [−]	irregular in shape, mostly aggregated	26.21 (PPy/EPS) 22.59 (PPy)				[113]
Cr(VI)	glycine doped PPy (PPy-gly)	SO ₄ ^{2−} zwitter ion of glycine	spherical particles (agglomerated) PPy: 153–538 nm PPy-gly: 150–250 nm		217.39–232.55	2.0	Langmuir	[114]
Cr(VI)	nanocomposite of Fe ₃ O ₄ with chitosan (CS) and PPy	Cl [−] SO ₄ ^{2−}	granular and irregular particles		105	2–4.5	Langmuir	[115]
Ni	polyethyleneimine/PPy	SO ₄ ^{2−}	Globular with an average particle size of 18–34 nm	17	1.756 (20 °C) 1.905 (60 °C)	4.5	Freundlich	[116]
Pb ²⁺ Cd ²⁺	rose leaf modified by PPy coating	Cl [−]	partially sponge-like structures with the pores		11.76 (for Cd ²⁺) 1.33 (for Pb ²⁺)	5.0 (for Cd ²⁺) 8.0 (for Pb ²⁺)	Freundlich	[117]
Mn(II) 17β-estradiol (E2), nitrate	PPy/corn cob immobilized with Zoogloea sp.					6.5		[118]

3.3. Sorption of Drugs

The polar nature of some pharmaceutical molecules induces their water solubility. As a result, metabolites and partially metabolized compounds are excreted via urine or feces, subsequently entering wastewater systems. Additionally, pharmaceutically active compounds (PACs) are often discarded as unused or expired products [120]. PACs are classified as contaminants of emerging concern (CECs) [121] or emerging pollutants known as the primary sources of environmental contamination, typically detected at low concentrations. Nevertheless, prolonged exposure to such contaminants may pose a potential risk to living organisms. Technologies traditionally used for water remediation based on filtration are not suitable for the removal of complex polar molecules. Hence, technologies based on advanced oxidation processes (AOPs) [122] or adsorption on appropriate solid materials such as activated carbon [123] were proposed. The AOP procedure involved high costs and significant energy consumption, failed to achieve complete pollutant mineralization, and could generate hazardous products or by-products; hence, the alternative methods were sought. Activated carbons were also tested in the PACs' removal; still, more adsorption data from this path are needed [124].

Salicylic acid (SA) plays a significant role in organic synthesis, while acetylsalicylic acid (ASA) serves as a commonly used non-steroidal anti-inflammatory drug with concentrations found in industrial wastewaters that are tens of $\mu\text{g/L}$ (SA) and $\mu\text{g/L}$ (ASA) [125]. The efficiency of SA and ASA removal through aqueous-phase adsorption strongly depends on the adsorbate's nature, as functional groups significantly influence the adsorption mechanism [126]. High efficiency was reported for the simultaneous removal of several pollutants, including SA, diclofenac (DCF), ibuprofen (IBU), ketoprofen (KETO), naproxen (NAPR), and tramadol (TRAM) from sewage [127] using cross-linked β -cyclodextrin (CD). Adsorption of SA, ASA, and atenolol from aqueous solutions was realized on clinoptilolite modified with sorbed metallic cations or natural clays (kaolin and bentonite, virgin or ion-exchanged using octadecyl dimethylbenzylammonium chloride) [128]. Details of the morphological and some physical properties of sorbents are depicted in Table 2. Chafai et al. [129] investigated the adsorption behavior of sodium salicylate (SA) and hexavalent chromium [Cr(VI)] onto polypyrrole (PPy), demonstrating the significant influence of experimental parameters such as the carrier mass-to-solution volume ratio, initial solute concentration, and pH. The retention of both adsorbates on polypyrrole occurred rapidly, while the extraction efficiency of sodium salicylate was unaffected by pH variations within the examined range [129]. In a related approach, cotton fabrics coated with polypyrrole and polyaniline were used as adsorbents for the salicylic acid and diclofenac removal [130]. The removal efficiencies exceeded 90% at pH 5.3 for DCF, while it was 70% at pH 4 for SA. The fast adsorption process reached equilibrium within 20–30 min. The better adsorption performance was noticed for PPy-coated fabrics, with adsorption capacities of 65 mg/g for DCF and 21 mg/g for SA. Higher total cohesive energy densities for adsorption on PPy-coated fabrics compared to PANI-coated ones validate this result, with the interaction mechanism involving Coulombic electrostatic attractions, non-Coulombic van der Waals forces, and π - π stacking interactions [130]. In a search for raw materials, another composite based on sunflower seed shell (SFS) coated with a small amount of polyaniline was reported by Albourine [131]. The experiment of the removal of sodium salicylate from water revealed the influence of parameters like adsorbent dose, initial concentration, pH of the solution, temperature, and contact time. The adsorption process was exothermic and spontaneous, according to thermodynamic parameters, with a dominant role of pristine concentration of analyte leading to an efficiency of 28.81 mg/g [131]. According to the literature, the adsorption efficiency of SA (mg/g) for a range of adsorbent types was 256.1

for modified $\text{SiO}_2/\text{Al}_2\text{O}_3$ [132], 238.3 for cross-linked PMADETA/PDVB [133], 210.5 for barely straw biochar [134], and 17.1 for dextrin-based polymer [135].

The adsorption of another drug, namely potassium diclofenac (KDCF), studied by Borges [136], involved the use of a pristine multiwalled carbon nanotube (pristine MWCNT) and the polypyrrole (PPy/MWCNT) composite as sorbents. Optimized procedure allowed for an adsorption of 93.48% and 94.98% for pristine MWCNT and PPy/MWCNT, while the surface area of PPy-modified sorbent increased markedly (from $277.5 \text{ m}^2/\text{g}$ to $541.2 \text{ m}^2/\text{g}$), leading to a maximum adsorption capacity change from 59.67 mg/g for the pristine MWCNT to 229.93 mg/g for the PPy/MWCNT [136]. The removal of potassium diclofenac (KDCF) and moxidectin (MOX) from aqueous solutions reported by Pires [137] utilized the PPy adsorbent characterized with a mesopore structure along with hydrophilic characteristics. Such a combination facilitated mass transfer and enhanced adsorption kinetics, leading to the high removal efficiency (95.26% for KDCF and 99.75% for MOX) granted by the sorption process at a pH of 6 [137]. The values of the adsorption efficiency (mg/g) of diclofenac for various types of adsorbents found in the literature spanned broadly from 217 for phenyl-phosphate-based porous organic polymers [138], 61 for poly(acrylonitrile-co-ethylene glycol dimethacrylate-co-vinylbenzyl chloride) [139], 9 (99.7%) for citrus-waste biomass to 4.15 (46.4%) for olive-mill residues [140], suggesting PPy-based composites as promising sorbents for DCF extraction.

A magnetic nanoparticle-enriched solid-phase extraction sorbent of polypyrrole/silica was applied for the extraction of sulfonamides in water samples [141]. Polypyrrole-coated silica achieved high extraction efficiency through π - π and hydrophobic interactions between the polymer and sulfonamides. Under optimized conditions, the method showed linearity in the range of 0.30 – $200 \text{ }\mu\text{g/L}$ for sulfadiazine and sulfamerazine, and 1.0 – $200 \text{ }\mu\text{g/L}$ for sulfamethazine and sulfamonomethoxine [141]. Another magnetic nanocomposite based on chitosan-polypyrrole (CS-PPy) was synthesized and tested in micro-solid phase extraction of naproxen [142]. Extraction efficiency of the CS-PPy magnetic nanocomposite elevated the value for the separation of components. Under the optimum condition, a linear calibration curve adhered to the range of 0.04 – $10.00 \text{ }\mu\text{g/mL}$ ($R^2 = 0.9996$), with an absolute recovery of 92% [142]. The magnetic nanocomposite of a polypyrrole—chitosan— Fe_3O_4 was also tested as a magnetic nanosorbent for the removal of carbamazepine (CBZ) from the wastewater (Figure 6) [143]. Under the optimal conditions, the sorbent revealed a high removal efficiency of 94.5% in 25 min in comparison to 65% for bare Fe_3O_4 magnetic nanoparticles. The CBZ adsorption on the composite was fitted with the Langmuir isotherm providing adsorption capacity (121.95 mg/g), which was higher than the one reported for CBZ sorbed on Fe_3O_4 -MAA (77.30 [144]) or expanded graphite amberlite XAD-7 (97.09 [145]) but lower than for fly ash-amended soil (213.74 [146]). The sorbent could be utilized up to five times for CBZ removal without a noticeable decrease in its adsorption efficiency [143].

An adsorbent composed of graphene oxide/covalent organic frameworks mixed with polypyrrole GO/COF-300 was described and used for the removal of indomethacin (IDM) and diclofenac (DCF) from aqueous solution [147]. The adsorbents showed an adsorption capacity of 115 mg/g for IDM and 138 mg/g for DCF after 60 min, with constant removal efficiency for drugs after eight cycles of regeneration [147]. Zeolite as a low-cost sorbent was supported with TiO_2 /polypyrrole nanoparticles to purify aqueous solution from rifampin and reactive orange 5 (RO5) [148]. The optimum contact time under ultrasonic-supported conditions for rifampicin and RO5 was 20 min, which allowed for reaching 94% and 88% removal efficiencies, respectively. Variation in optimal pH value for the impurities removal (two for dye and five for rifampicin) stressed different behavior in terms of interactions with the sorbent's functional groups [148]. Also, the association of TiO_2 with polyaniline was verified as an adsorbent aimed to degrade organic pollutants [149]. Inorganic oxides such as titanium dioxide and zinc oxide function as active

photocatalysts, and hence can extend the impurity removal process with a contribution of photocatalytic decomposition [32]. The light absorption of TiO_2 -based composite was expanded to the visible range, as proved by a decrease in gap energy values to 2.5 eV, which enabled the degradation of salicylic acid in the presence of simulated sunlight [149]. Similar semiconductive silver molybdate (Ag_2MoO_4) with PPy was prepared as a nanocomposite for the degradation of pollutants and heavy metals, e.g., methylene blue (MB) and Cr(VI), as well as antibiotic ciprofloxacin (CIP) [150]. The system was studied in terms of its activity in photocatalysis and electrochemical sensing capability. The nanocomposite proved its utility as an efficient photocatalyst in the degradation of MB (99.9%), Cr(VI) (99%), and CIP drug (99.8%) within 10 min. One-pot synthesis of a nanocomposite composed of silver manganite incorporated PPy (AMN-PY) was proposed by Abinaya et al. [151]. The composite exhibited activity in the photocatalytic degradation of methyl orange dye (99.6%) and chloramphenicol (CAP) drug (98.9%) within 30 min. PPy and AMN exposed to sunlight generated excited electrons and holes that reacted with water to form reactive radicals, initiating pollutant degradation (Figure 7). Silver ions enhanced the photocatalytic process by serving as electron reservoirs, while PPy particles prolonged electron-hole separation and reduced recombination rates, thereby elevating the overall degradation efficiency [151].

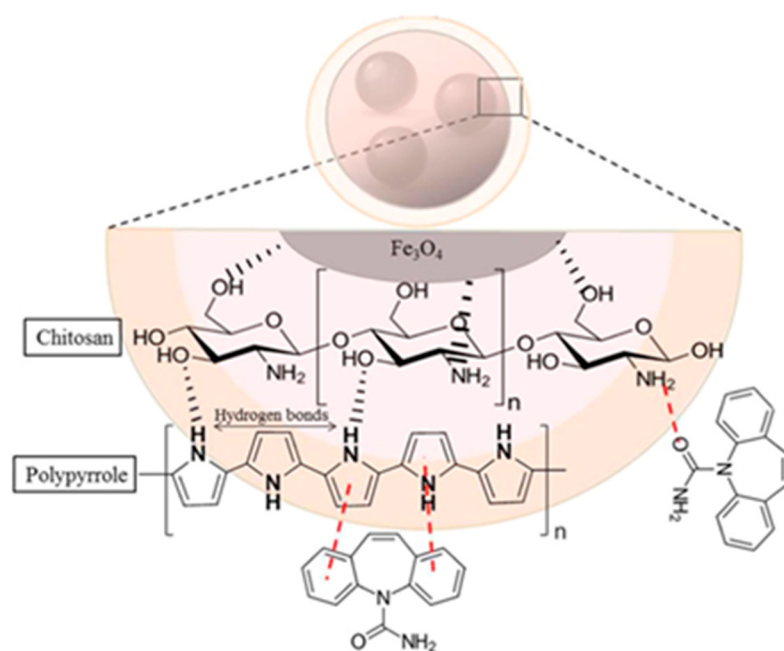


Figure 6. Schematic representation of the PPy-CS- Fe_3O_4 nanocomposite structure with proposed interactions (reproduced with permission from [143]; published by Elsevier, 2021).

A similar approach based on photocatalytic activity was proposed in studies of SA removal with polypyrrole- TiO_2 composite [152], diclofenac removal with polypyrrole-doped reduced graphene oxide [153], clofibric acid (CLA), diclofenac (DCL), and indomethacin (IND) removal [154], where photocatalytic degradation paths were employed. As presented, PPy demonstrates strong performance in the removal of pharmaceutical compounds from aqueous systems. Its π -conjugated polymer backbone, coupled with heteroatoms and functional groups such as nitrogen-containing sites, enabled multiple binding mechanisms including π - π interactions, hydrogen bonding, and electrostatic attraction with aromatic or polar pharmaceutical molecules. As a result, PPy-based materials achieve high removal efficiencies for a wide range of pharmaceuticals as listed previously. When compared with conventional sorbents such as activated carbon, zeolites, silica gel, and metal oxides, PPy exhibits several key advantages. It offers a chemically versatile and electrochemically responsive surface,

allowing for tunable interactions with pharmaceutical compounds. Its selectivity can be enhanced through doping strategies (e.g., using anionic dopants like sulfonates or phosphates) or by forming composite materials with increased surface area and targeted functionalities. One of PPy's most compelling advantages lies in its regeneration mechanism. Unlike activated carbon or metal oxides, which often require chemical reagents or high-temperature treatment for regeneration, PPy can be reversibly regenerated electrochemically by switching its redox state. The regeneration strategy not only extends the lifespan of the sorbent but also allows for the potential recovery of high-value pharmaceutical compounds.

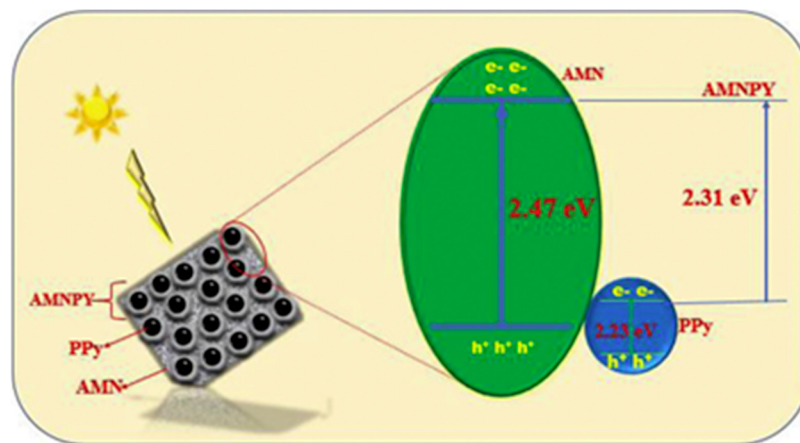


Figure 7. Schematic representation of the possible charge transfer mechanism of AMNPY (reproduced with permission from [151]; published by Elsevier, 2020).

Table 2. Comparison of the synthetic and physicochemical details of PPy-based sorbents aimed at drug removal.

Sorbed Drug	Description	pH/efficiency [%]	S BET [m ² /g]	Adsorption Capacity [mg/g]	Equilibrium Times	Cycles of Regeneration [no]	Ref.
ketoprofen (KETO)	cross-linked β -cyclodextrin (CD)	4.2	7.31	API 162.20	60 min		[127]
salicylic acid, acetylsalicylic acid, atenolol	clinoptilolite modified with sorbed metallic cations or natural clays (kaolin and bentonite, pure or ion-exchanged by octadecyl dimethylbenzyl ammonium chloride)			0.65–3.5 $\times 10^{-5}$ mol/g for SA 1.1–2.1 $\times 10^{-5}$ mol/g for ASA			[128]
sodium salicylate	PPy powder	largely unaffected by pH					[129]
salicylic acid, diclofenac	cotton fabrics/PPy or PANI	90% at pH 5.3 for DCE, 70% at pH 4 for SA		65 mg/g for DCF 21 mg/g for SA	20–30 min		[130]
sodium salicylate	sunflower seed shell (SFS)/PANI composite	6/90% for 1 g/L		28.81 mg/g	60 min		[131]
potassium diclofenac	pristine multiwalled carbon nanotube (pristine MWCNT)/PPy	93.48% for pristine MWCNT 94.98% for PPy/MWCNT	277.5 (MWCNT) 541.2 (PPy/MWCNT)	59.67 mg/g (pristine MWCNT) 229.93 mg/g (PPy/MWCNT)			[136]
potassium diclofenac (PD) moxidecin (MOX)	PPy	pH 6/95.26% for PD, 99.75% for MOX	27.22	221.23 for PD 87.46 for MOX	20 min	7	[137]
diclofenac (DCF) salicylic acid	cotton fabrics coated with PPy or PANI	pH 5.3 for DCE, pH 4 for SA/ 90% for DCE, 70% for SA		PPy-coated fabrics: 65 for DCF, 21 for SA	20–30 min	3	[139]
sulfonamides	a magnetic nanoparticle/PPy/silica	7.0			2–20 min	16	[141]
naproxen	magnetic nanocomposite chitosan-polypyrrole (CS-PPy)	5.0/92%			15 min		[142]
carbamazepine (CBZ)	PPy/chitosan—Fe ₃ O ₄	94.5% for PPy/CS/Fe ₃ O ₄ , 65% for bare Fe ₃ O ₄		121.95	25 min	5	[143]
indomethacin (IDM), diclofenac (DCF)	graphene oxide/covalent organic frameworks/PPy (GO/COF-300/PPy)	72% for DM, 68% for DCF		115 (for IDM) 138 (for DCF)	60 min	8	[147]
Rifampin, reactive orange 5 (RO5)	zeolite supported with TiO ₂ /PPy nanoparticles	pH = 5 for Rifampin, pH = 2 for RO5/94% for Rifampin, 88% for RO5			20 min (ultrasonic-supported conditions)		[148]
salicylic acid	TiO ₂ /PANI	71% for T-Pani-6 (100:1), 75% for T-Pani-7 (200:1), 34% for TiO ₂			5 h (of white light illumination)		[149]
ciprofloxacin (CIP) methylene blue (MB), Cr(VI)	silver molybdate (Ag ₂ MoO ₄)/PPy	99.8% for CIP, 99.9% for MB, 99% for Cr(VI) photocatalytic degradation			8–10 min		[150]
chloramphenicol (CAP), methyl orange (MO)	silver manganite/PPy (AMNPY)	99.6% for MO, 98.9% for CAP photocatalytic degradation			30 min		[151]

3.4. Sorption of Dyes

The extensive use of dyes in industries such as textiles, food, cosmetics, paints, and printing significantly contributes to aquatic pollution. Dyes can be classified as inorganic or organic, with the latter predominantly used in the textile and food industries [155]. Effluents containing dyes pose serious environmental and health concerns, including toxicity, mutagenicity, and disruptions in photosynthesis caused by dye contamination [156]. Reactive Red 120, commonly applied in textile dyeing, has been reported to cause chronic skin inflammation and bronchial asthma [157]. Sulfur Black, primarily used for dyeing cellulosic fibers, is known to cause skin and eye irritation in humans [158]. An anthraquinone dye, namely, Vat Green 3, utilized in the coloring and printing of cotton fibers and cotton blends, exhibits high toxicity to marine life [159]. Acid Violet 7, used as a colorant for wool, silk fibers, and beauty products, has been identified as a potent human carcinogen [158]. Similarly, Acid Orange 7, commonly employed as a hair dye, has been found to cause irreversible morphological damage to organic tissue [160]. Eriochrome Black-T (EBT) dye presents significant toxicity and resists removal from water sources due to its strong photostability [161]. EBT dye alters the activity of metabolic and antioxidant enzymes upon entering the human body and activates hazardous metabolites that stimulate free radical production in exposed human cells [162]. Addressing these issues is of critical importance, with several available solutions like removal techniques (Table 3), including the utilization of polypyrrole-based composites as adsorbents, particularly for the treatment of textile wastewater.

Table 3. Dye removal techniques limitations (produced based on [163,164]).

Method	Shortcomings
Adsorption	Multiple parameters govern the process
Flocculation	Development of sludge precipitates
Electrolytic precipitation	Demands significant processing time
Electrochemical oxidation	High operational cost driven by electrical energy usage
Ion exchange	Effective for selected dyes
Fenton process	Excessive generation of anionic species
Membrane filtration	Characterized by limited stability and high cost
Phytoremediation	Provides a non-permanent remediation solution
Bioremediation	Inhibits microbial growth and activity
Photocatalytic degradation	Requires a light source, which contributes to increased operational costs

Polypyrrole by itself serves in adsorption applications because nitrogen atoms within its polymer chains actively chelate with heavy metals and dye effluents through their positive charges in the polymeric matrix. Additionally, oppositely charged ions are incorporated into the polymer chains to maintain overall charge neutrality. These distinctive properties establish PPy as an effective adsorbent [165,166]. Moreover, the enhanced efficiency of the process is sought in the development of hybrid PPy composites that provide a synergistic effect between PPy and the component material. Among materials utilized to form a composite with PPy, one can find sawdust (SD), rice husks, chitosan, cellulosic biomass, and sugarcane bagasse. The SD-based composite with PPy was studied by Palanisamy [167]. The oxidative polymerization with FeCl_3 as oxidant led to composite formation. The powder product used as an adsorbent for dyes, e.g., AO 10 (Acridine Orange 10), proved to be efficient by removing 243.9 mg/g of dye in a process that followed pseudo-second-order kinetics. This result indicated an ion exchange mechanism, where electrostatic interactions between the positively charged active sites of PPy and the negatively charged AO10 dye anions predominantly governed the adsorption process [167].

PPy-rice bran (Rb) biocomposites were tested for adsorption studies of malachite green (MG) dye [168]. The composite utility for MG dye adsorption was verified in terms of the optimum temperature (50 °C), contact time (1 h), and adsorbent dose (0.05 g/L) at acidic pH. The adsorption process followed a pseudo-second-order mechanism [168].

Agrarian wastes are a source of composite components for efficient dye removal owing to their low price, biodegradability, and accessibility. Among them, one can find fruit peel-based biomass, e.g., pineapple, orange, pitahaya, pumelo, or garlic peels, nut and seed shell-based biomass, e.g., coconut, groundnut shells or peanut hulls, and fibrous crop byproducts, e.g., sugarcane bagasse or bamboo shells [169]. Wang [170] demonstrated the potential of Chinese yam peelings (CPy) as sustainable adsorbents for wastewater treatment. By integrating PPy with CPy, the sorption process achieved 86% removal of Congo Red (CR) dye from a 100 ppm solution within 2 h using 10 g L⁻¹ of adsorbent at 45 °C. The adsorbent maintained its performance after regeneration and enabled multiple reuse cycles without significant loss of efficiency [170]. Another commonly used component of PPy-based composite is chitosan, which is an organic, biodegradable polysaccharide-based macromolecule. Its abundant availability, along with surface-exposed -NH₂ and -OH functional groups, enables efficient adsorption of heavy metals and dyes [171]. Chitosan adsorbs onto the PPy surface and functions as a size stabilizer during polymerization, preventing the agglomeration of PPy particles and enhancing adsorption performance, which leads to enhanced productivity, shortened time, and effortless recovery of the adsorbent. Such a PPy/chitosan composite was applied in the removal process of acid red (AR 18) dye [172]. Glacial acetic acid CH₃COOH and APS (NH₄S₂O₈) were used to prepare the material. Moreover, 98.71% of dye was removed in half an hour in a pseudo-second-order reaction, and the adsorbent was reusable for four cycles [172]. Biocomposites composed of chitosan, starch, polyaniline, and polypyrrole with sugarcane bagasse were prepared and tested for the removal of the Acid Black-234 dye [173]. Some kinetic data of biosorption processes were described with the pseudo-first-order kinetic model (SB, PAn/SB, PAn/Ch, PPy/St, and PAn/St), while the other with the pseudo-second-order kinetic model (PPy/SB and PPy/Ch). This implied that, for the first set of sorbents, it is a surface-controlled process with a dominant role of physical adsorption (physisorption); meanwhile, for the second set, it was rather a chemisorption-dominated process controlled by chemical bonding between adsorbate and adsorbent functional groups. The biosorption of the dye followed the Langmuir isotherm model, with 100 PPy/SB exhibiting the highest adsorption capacity, while thermodynamic analysis confirmed the exothermic nature of the process [173].

The presence of polymer fibers like cellulose at the PPy preparation stage led to a shrinking of the size of the final product and stabilized it with the presence of functional groups. In the field of adsorption phenomena, these groups increased the possibility of interaction between the adsorbent and active sites of dye particles, provoking enhanced adsorption activity [174]. A PPy/nanocellulose composite studied by Shahnaz [175] served as a sorbent for the removal of CR dye and Cr ions from a binary mixture. Nanocrystalline cellulose (NCC) obtained from the acid hydrolysis of cellulose fibers was known for its high specific strength and modulus, as well as high surface area [176]. The synthesized NCC was mixed with PPy formed in oxidative polymerization with the use of APS. Batch adsorption of the CR dye from a mixture containing Cr(VI) ions demonstrated 85% dye removal under optimal conditions (pH equal to 2, initial concentration of 30 ppm, adsorbent dose of 0.02 g, temperature of 30 °C), despite the presence of Cr ions as secondary contaminants [175]. Also, the PPy/SiO₂ nanocomposite was verified as a novel adsorbent for capturing AO7 dyes [177]. Silica nanoparticles were used owing to their very small size, which led to properties like low toxicity and high rigidity, with a high surface-to-volume ratio, urgent from an adsorption point of view [178]. Parameters' optimization involved adsorbent dosage,

shaking speed, dye concentration, and temperature. The adsorption process itself is composed of both surface adsorption and particle diffusion. The highest values of R^2 were noted, and the Langmuir isotherm model proved the occurrence of monolayer coverage of AO7 molecules on the PPy/SiO₂ surface as highly probable. The mean adsorption energy (E) was found to be below 8 kJ/mol, indicating that the adsorption process was driven by interactions like hydrogen bonding, hydrophobic interactions, electrostatic attractions, and specific interactions between the adsorbate and the PPy/SiO₂ composite [179]. Another silica-based SBA-15/polypyrrole composite with a mesoporous hexagonal structure served as a sorbent for the uptake of dyes (including sunset yellow, indigo carmine, titan yellow, and orange G) from aqueous solutions [180]. The efficiency for all the dyes exceeded 90%, with the maximum adsorption capacities found as 78.74 mg/g (sunset yellow), 83.33 mg/g (indigo carmine), 78.74 mg/g (titan yellow), and 106.38 mg/g (Orange G). The SBA-15/PPy composite was prone to recycling, with 90% removal efficiency reported for three consecutive cycles [180]. Polypyrrole-decorated bentonite magnetic nanocomposite (MBnPPy) was prepared by Ahamad in a two-step process [34] and tested for the removal of both anionic methyl orange (MO) and cationic crystal violet (CV) dyes from contaminated water. Bentonite, being a type of clay, exhibits chemical and mechanical stability, possesses a large surface area, and demonstrates a high swelling capacity. The presence of PPy and magnetic nanoparticles enhances their conductivity, surface area, regeneration potential, and adsorption capacity and selectivity. Setting the sorption experiment under optimal conditions, including an initial dye concentration of 100 mg/L, MBnPPy dosage of 2.0 g/L, equilibrium times of 105 min for MO and 120 min for CV, pH adjusted to 5.0 for MO and 8.0 for CV, and a constant temperature of 303.15 K, resulted in monolayer adsorption capacities of 98.04 mg/g for MO and 78.74 mg/g for CV. Regeneration ability was satisfactory for up to five adsorption–desorption cycles [34]. An organometallic composite composed of polypyrrole-encapsulated zinc oxide (PPy/ZnO) was tested for the elimination of Eriochrome black-T (EBT) dye from water [181]. Parameters like specific surface area and pore volume of the nanocomposite were reported as 28.726 m²/g and 0.086 cm³/g. Maximum dye removal efficiency (94%) was denoted for the optimal condition, including a solution pH of 4.0, 1.0 g/L of nanocomposite dose, 30 min contact time, and 50 mg/L of initial EBT dye concentration, which led to the maximum EBT dye adsorption capacity of 277.78 mg/g. In the process, mainly the hydrogen bonding, π – π interactions, and electrostatic attraction were responsible for the sorption act, while five regeneration cycles occurred successfully with a 20% decrease in dye removal efficiency. The investigation also examined how real water or wastewater conditions and the presence of multiple co-existing ions affected adsorption efficiency [181]. Other CPs were also tested as sorptive candidates, including polyaniline/zinc oxide composite for methyl orange adsorption [182]. Hierarchical porous zinc oxide microspheres were utilized for Congo red removal (maximum adsorption amount of 334 mg/g) [183], while zinc oxide nanoparticles loaded on activated carbon served to remove malachite green from the aqueous solution (maximum adsorption capacity was 322.58 mg/g) [184]. The findings reveal comparable sorption capacities among PPy-based sorbents, while also highlighting scope for procedural enhancement. Polypyrrole-coated cobalt ferrite nanocomposite effectively removed the cationic dye Basic Blue 3, achieving a maximum adsorption capacity of 130.1 mg/g under optimal conditions along with efficient five desorption-adsorption cycles operation [185]. PPy exhibits a strong affinity toward synthetic dyes, making it highly effective for treating dye-contaminated industrial effluents, especially from textile, paper, and pharmaceutical industries. The presence of π -conjugated bonds and nitrogen-containing functional groups in the PPy backbone enables multiple interaction pathways with dye molecules, including π – π stacking, electrostatic forces, hydrogen bonding, and ion exchange with dopant anions.

These interactions allow PPy to effectively adsorb dyes as presented above, across a range of pH and ionic strength conditions. In comparison to conventional sorbents, PPy offers several advantages. Activated carbon is widely used due to its large surface area and porosity; however, its interactions with dyes are largely nonspecific, and its performance deteriorates in the presence of surfactants, salts, or organic matter [186]. Moreover, regeneration typically requires high-temperature or chemical treatment, which can degrade the structure and limit reusability. Zeolites, while effective for certain dye classes through ion-exchange mechanisms, are generally more selective toward small, cationic dyes and are less effective for large or anionic molecules [187]. Silica gel adsorbs dyes primarily through physical interactions and surface hydroxyl groups [188] but lacks selectivity and exhibits limited capacity under high dye concentrations or variable pH. PPy electrochemical tunability allows for the dynamic modulation of surface charge, namely oxidized PPy, which can preferentially attract anionic dyes, while reduced PPy can facilitate desorption. Cationic and anionic dyes interact differently with PPy due to electrostatic forces (e.g., the anionic azo dye Acid Red 26 forms stable aggregates with cationic surfactants, highlighting the importance of charge interactions in adsorption processes [189]). The adsorption of reactive dyes is also influenced by the presence of amino groups on polymeric adsorbents, which enhance the interaction with negatively charged dyes [190]. The size of dye molecules affects their adsorption capacity. Smaller dyes can penetrate the polymer matrix more easily, as seen, e.g., with Biebrich scarlet and Eosin Y, which exhibited high removal efficiencies due to their smaller molecular sizes [191]. The surface morphology of PPy, influenced by the dye structure, can lead to different adsorption behaviors, as demonstrated by the varying conductivity and doping levels of PPy synthesized with different azo dyes [192]. Hydrophobic interactions play a crucial role in the adsorption of dyes onto PPy. Dyes with hydrophobic characteristics tend to adsorb more effectively due to favorable interactions with the polymer matrix [192]. π - π stacking interactions between the aromatic structures of dyes and the PPy matrix enhance adsorption, as evidenced by the structural studies that reveal the geometry of dye aggregates [189]. Synthetic strategies also focus on minimizing agglomeration to facilitate molecular ingress, thereby increasing the accessible surface area and enhancing active site availability. While these factors significantly enhance dye adsorption on PPy, it is essential to consider that the overall efficiency can vary based on environmental conditions and the specific characteristics of the dyes and polymers involved.

3.5. Bacteria Removal

Another interesting area of application for PPy composites is the decontamination of water and surfaces. Microbial contamination of water was reduced with the aid of polypyrrole magnetic nanoparticles ($\text{Fe}_3\text{O}_4/\text{PPy}$ NPs) [193]. The study demonstrated over 50% bacterial adsorption at a nanoparticle dosage of 100 $\mu\text{g}/\text{mL}$ under a magnetic field. Additionally, near-complete photothermal sterilization ($\sim 100\%$) occurred after 10 min of NIR exposure, effectively inactivating *E. coli* and *S. aureus* in a water system. Such behavior was observed in both the adsorption capability of nanoparticles governed by electrostatic interaction between NPs and bacterial surfaces and high photothermal conversion efficiency of NPs (Figure 8), resulting in an increase in the temperature of the bacterial suspension beyond the threshold for bacterial survival [193]. Copper-modified polypyrrole films disinfected well water contaminated with *E. coli* [194] in a lab-scale continuous flow system. The bacteria-killing process was described by a first-order kinetic law with a more enhanced process for increased flow rate. Re-inoculation and Cu-recharging tests produced bactericidal effects comparable to those of freshly prepared electrodes [194]. The poly(vinyl alcohol) (PVA)-stabilized polypyrrole nanoparticles (PVA-PPy NPs) mixed with tannic acid

(TA) [195] exhibited photothermal properties under 808 nm near-infrared irradiation. This behavior induced bacterial death through the hyperthermal effect, resulting in *S. aureus* and *E. coli* survival rates of 1.66% and 2.78%, respectively, on the surfaces. Moreover, the covered surface efficiently prevented the formation of early-stage biofilm under NIR irradiation [195].

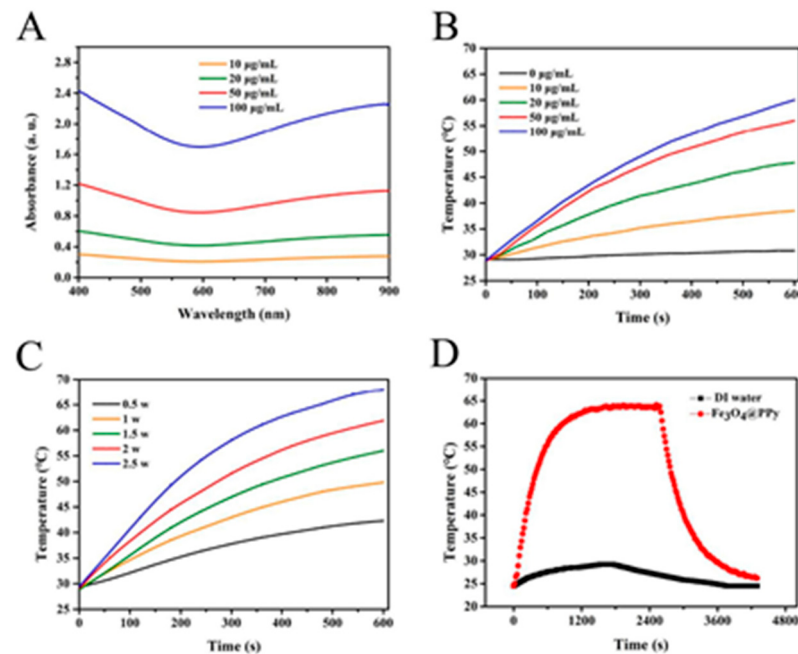


Figure 8. (A) Fe₃O₄@PPy nanoparticles (NPs) UV-vis spectra, (B) temperature dependence for different concentrations of NPs, (C) temperature dependence NPs with various laser power densities, and (D) photothermal heating curves of the NPs and DI water (reproduced with permission from [193]; published by Elsevier, 2021).

Another path of application of PPy is enhanced water quality monitoring with a rapid but significant analyte signal [196]. In this field, electrochemically active biofilm (EAB)-based biosensors attract intensive research interest because they generate electrical signals applicable for water quality monitoring [197]. EAB, composed of electroactive bacteria embedded in a matrix of extracellular polymeric substances, attaches to carriers such as minerals or electrodes [198]. In this configuration, biosensors function as early-warning systems for pollutants, as interactions with incoming toxicants alter the stable electrical signal generated by the EAB biosensor [199]. In the work of Liang [196], the artificial EAB composed of *Shewanella oneidensis* MR-1 encapsulated in sodium alginate (SAI) was prepared with PPy as a conductive medium to facilitate the extracellular electron transfer (EET). The results showed that a small amount of PPy (0.125:1 mass ratio of PPy:SAI) was the most effective, as at this configuration, enhanced conductivity increased sensitivity, while mass transfer experienced a concurrent decline [196].

3.6. Membrane Separation

In purification procedures, besides AOP and adsorption methods, membrane technology is also useful [200,201]. Membrane-based treatment technologies offer an attractive approach for remediating polluted water resources due to their high contaminant selectivity, capacity for continuous and automated operation, and minimal spatial and energy demands [202]. Bearing in mind its high cost, the contaminants can be obtained in a disposable form derived from the significantly reduced volume [203]. Membranes are designed to provide proper permeability and porosity that will improve both flux and economics [204].

Within membrane technologies, ultrafiltration (UF) finds extensive application in protein purification, bacterial removal, food product fractionation, and oil–water separation [205]. From a materials standpoint, ultrafiltration (UF) membranes are commonly fabricated from specialized polymers such as poly(vinylidene fluoride) (PVDF), poly(ether sulfone) (PES), polyacrylonitrile (PAN), and polysulfone (PS), owing to their exceptional chemical stability, high efficiency, robust mechanical performance, and superior heat resistance. Biofouling poses a significant challenge, obstructing the sustainable advancement of membrane separation technology. The wetting property dictates the path of the process - a hydrophilic feature brings resistance to fouling during filtration, still a hydrophobic nature is more desired at the cleaning stage. Hence, the membrane materials capable of changing the wettability are intensively sought. Moreover, perspective features like selective pollutant adsorption and catalytic detoxification might be induced by surface engineering outlook. Nanofiltration (NF) membranes are thin-film composite [206], which act as the active layer, and are commonly supported by polysulfone (middle) and polyester (bottom) sublayers. The separation process is a sieving process that depends on size exclusion, surface hydrophilicity, and surface charge.

Microwave (MW)-assisted in situ growth incorporated carbon nanotubes (CNTs) into commercial polyamide (PA) nanofiltration membranes, specifically NF90 and NF270. PPy and a ferrocene catalyst facilitated the growth process by increasing the local temperature and inducing the growth of CNTs [207]. The NF270-PPy-CNT membrane proved its usability in ion rejection tests with a 14% increase in ion rejection. Analysis based on the Spiegler–Kedem model revealed that it was diffusive transport that contributed mostly to enhanced desalination [207]. Hudaib [208] employed phase inversion to fabricate a poly(vinylidene fluoride) (PVDF)/multiwalled carbon nanotube (MWCNT)/polypyrrole (PPy) ultrafiltration membrane designed for crude oil removal from refinery wastewater. The characterization of material revealed that the pristine PVDF membrane permeability can be elevated 17-fold by adding MWCNT (0.05%), while the membrane removed approximately 99.9% of crude oil. The hydrophilic surface of the membranes was verified as a consequence of the hydrophilic nature of the nitrogen component [209], while the membrane's rough surface structure enhanced wettability and reduced the contact angle (CA) [210]. The CAs of the modified membranes decreased from 85° in the unmodified form to 65°, indicating enhanced hydrophilicity resulting from the MWCNT/PPy complex interaction. This interaction also accelerated solvent (DMF)-non-solvent (water) diffusion, promoting the development of a more porous membrane structure and increasing porosity from 65.6% to 91.8%. A high rejection efficiency of crude oil was noted for the studied membranes, with a value exceeding 99% for the modified ones. Another method aimed at water purification was described by Bao [211], which used photo-Fenton-active (PFA) membranes. In the system, photocatalysis and Fenton reaction were enhanced for effective solar-driven water treatment through the use of generated hydroxyl radicals ($\cdot\text{OH}$). The proposed PFA membrane was doubly functionalized with PPy, which acted like a photothermal catalyst, and zerovalent iron nanoparticles that were found to act as a photo-Fenton catalyst. High efficiency of organic dye molecules removal was reported (>97%) with the enhanced anti-fouling and self-cleaning properties of the membrane. Also, the applicability of the membrane for oil-emulsion filtration experiments was validated [211]. Membrane antifouling procedure was proposed by Liang [212], where a stainless steel mesh sulfosuccinate-doped polypyrrole composite (SSM/PPy(AOT)) was tested. The nature of the surface (its wettability) was converted by switching a negative or positive potential (Figure 9). The reduced state increased hydrophilicity by exposing the sulfonic acid groups of AOT at the surface. The filtration process led to resistance to fouling in the hydrophilic state, while the membrane was easier to clean in the hydrophobic state. SEM image analysis

showed that the local membrane surface exhibited increased roughness in the oxidation state and became smoother in the reduction state [212].

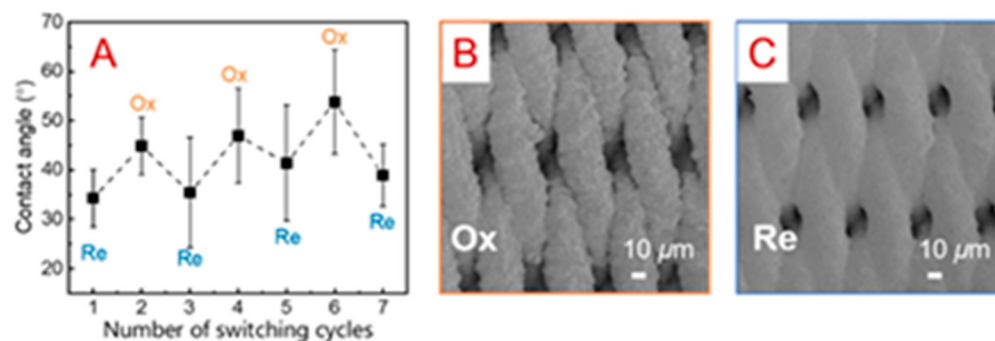


Figure 9. (A) Changes in measured water contact angles during membrane transition between oxidation (Ox) and reduction (Re) states; (B) SEM micrograph of the membrane in the oxidation state; and (C) SEM micrograph of the membrane in the reduction state (reproduced from [212] under the terms and conditions of the Creative Commons Attribution (CC BY) license (<https://creativecommons.org/licenses/by/4.0/>)).

Zhu [213] enhanced membrane antifouling performance and selectivity by employing polypyrrole-dodecylbenzene sulfonate (PPy-DBS) to create a membrane with tunable pores. Applying an external redox potential enabled in situ tuning of membrane pores, as ion insertion or expulsion altered the volume of PPy-DBS. Applying a positive potential enlarged the membrane pore size, whereas applying a negative potential reduced it. Adjusting the membrane pore size in combination with a cleaning step mitigated membrane fouling, offering an effective fouling control strategy.

4. Perspective

The adjustable sorption capacity of PPy-based composites presents a significant opportunity for the development of modern materials for water purification and detoxification. Considerable efforts are directed towards increasing the specific surface area and precisely controlling the size and extent of porosity, aimed at achieving a higher degree of contaminant removal and detoxification efficiency. Given the growing severity of water contamination, the urgent need for wise and efficient solutions is apparent. To meet these demands, it is essential to optimize synthetic protocols, carefully tuning both composition and procedure to match the requirements of programmed applications. A critical challenge remains in balancing material properties and practical usability, ensuring that performance enhancements do not compromise operational stability or scalability. Also, the importance of recovery and reusability of PPy-based materials must be addressed to promote sustainable practices. Moreover, these composites offer a wide range of applications in membrane separation technologies, where their integration enables an elegant and effective separation mechanism. The fabrication of thin membranes with improved antifouling properties represents a promising application path. The prospective development of nanocomposites based on PPy is expected to bring solutions for complex environmental challenges. Combined advantages in terms of selectivity, reusability, and functionality in complex matrices position PPy as a multifunctional sorbent platform for integrated removal of both inorganic and organic contaminants.

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