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Green polymeric nanomaterials for the photocatalytic degradation of dyes: a review

Shrabana Sarkar¹ · Nidia Torres Ponce² · Aparna Banerjee³ · Rajib Bandopadhyay¹ · Saravanan Rajendran⁴ · Eric Lichtfouse⁵

Abstract

Pure and drinkable water will be rarer and more expensive as the result of pollution induced by industrialisation, urbanisation and population growth. Among the numerous sources of water pollution, the textile industry has become a major issue because effluents containing dyes are often released in natural water bodies. For instance, about two years are needed to biodegrade dye-derived, carcinogenic aromatic amines, in sediments. Classical remediation methods based upon physico-chemical reactions are costly and still generate sludges that contain amine residues. Nonetheless, recent research shows that nanomaterials containing biopolymers are promising to degrade organic pollutants by photocatalysis. Here, we review the synthesis and applications of biopolymeric nanomaterials for photocatalytic degradation of azo dyes. We focus on conducting biopolymers incorporating metal, metal oxide, metal/metal oxide and metal sulphide for improved biodegradation. Biopolymers can be obtained from microorganisms, plants and animals. Unlike fossil-fuel-derived polymers, biopolymers are carbon neutral and thus sustainable in the context of global warming. Biopolymers are often biodegradable and biocompatible.

Keywords Biopolymer · Nanomaterial · Photocatalyst · Dye degradation

Introduction

According to the latest report of World Health Organization (WHO), approximately 844 million people worldwide lack the access to basic drinkable water (Wutich et al. 2019). Waterborne pathogens in the form of disease-causing bacteria, virus or protozoa spread many diseases including cholera, typhoid, hepatitis, giardia and COVID-19 (Sharma et al. 2020). Unsafe water causes epidemics in developing countries due to improper management of water pollution (Alhamlan et al. 2015). Organic pollutants in wastewater are potentially harmful for all living organisms. Regular consumption of untreated or poorly treated waters induces carcinogenesis or prolonged illness in humans and other animals (Sarkar et al. 2017). As a consequence, wastewater remediation and water recirculation are now the major research focus (Wen et al. 2019; Karimi-Maleh et al. 2020a). Particularly, the regulation of water contamination and recycling the wastewater should be improved in drought-affected countries (Gholami et al. 2019).

The negative health effects of water pollution are a major source of mortality worldwide (Wang and Yang 2016; Sarkar et al. 2017). In particular, water pollution has historically

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impacted food safety (Lu et al. 2015). The textile industry represents a threat when dye effluents are released into water bodies. Textile wastewater contains various contaminants such as synthetic azo dyes. Therefore, environmental legislation commonly obligates textile factories to treat effluents before discharge (Yaseen and Scholz 2019). Dye effluents are high in colour, pH, suspended solids (SS), chemical oxygen demand (COD), biochemical oxygen demand (BOD) (Yaseen and Scholz 2016), metals (Sharma et al. 2007; Sekomo et al. 2012), temperature (Dos Santos et al. 2007; Shah et al. 2013) and salts (Yaseen and Scholz 2019). Synthetic textile dyes are often recalcitrant and carcinogenic by nature due to the presence of $-N=N-$ bond (Singh et al. 2015). Those dyes mainly consist of complex aromatic structures that are hardly biodegradable.

Wastewater treatment involves a step of physicochemical fractionation, which separates hydrophilic and hydrophobic matter (Kim and Yu 2005). Techniques and adsorbents used for wastewater treatments have been recently compared (Crini and Lichtfouse 2019; Crini et al. 2019a). Methods for the treatment of dye-contaminated waters include reverse osmosis, coagulation, flocculation, ion exchange, activated carbon adsorption, advanced oxidation, ozonation, photocatalysis, Fenton process, photo-Fenton, electrochemical oxidation (Lade et al. 2015) and filtration (Singh et al. 2015). These processes are often expensive and generate amine residues found in sludges after treatment. Alternatively, semiconductors such as titanium dioxide and zinc oxide have shown excellent photocatalytic activity due to a positive band position that develops more electrons and holes under UV light (Fujishima and Honda 1972; McLaren et al. 2009; Xu et al. 2019). Recently, the photocatalytic capacity has been improved by modifying material surfaces using metal doped, non-metal doped and coupled systems (Gnanasekaran et al. 2017, 2018; Sulaiman et al. 2018; Kuo et al. 2019). Composite or coupled systems are now used for solar cells, opto-electronics, bio-electrochemical sensors, electro-oxidation and disinfection (Li et al. 2011; Devi and Kavitha 2013; Rokhmat et al. 2017; Karimi-Maleh et al. 2019; Karimi-Maleh and Arotiba 2020; Karimi-Maleh et al. 2020b, c). Here, we review green polymeric nanomaterials for photocatalytic dye degradation with special emphasis on recent developments, biopolymers and applications in wastewater remediation.

Synthesis of biopolymeric nanomaterials

Conventional methods to synthesise polymeric nanomaterial employ chemical compounds that may cause environmental toxicity later due to their long-term stability. By contrast, biopolymers are usually composed of safe monomers and are carbon neutral for the climate. Biopolymers facilitate the

synthesis of nanomaterials because biomass morphology is often structured at the nanolevel. Biopolymers are found in various organisms such as plants, algae, fungi, bacteria and animals. Macromolecules include starch, alginate, chitosan, dextran and chitin (Fig. 1). Chitosan, starch, dextran and cellulose are polysaccharides derived from plants and microbial biofilms, and these biopolymers are common for nanomaterial synthesis (Banerjee and Bandopadhyay 2016; Farshchi et al. 2019; Kolangare et al. 2019). In particular, chitosan has been used for dye removal and wastewater treatment (Crini et al. 2019b; Lichtfouse et al. 2019).

Biopolymers are unique in composition and have various physiological properties. Biopolymeric nanomaterials can be formed by attachment of metals to biopolymers. In particular, biopolymers form molecular capsules by intramolecular hydrogen bonding. For example, starch may incorporate metals or metal oxide, thus forming polymeric nanocomposites. Chitosan is also used for nanotechnology-related applications due to its wide compatibility (Vanaamudan et al. 2018; Morin-Crini et al. 2019). Silver (Ag) can be incorporated within starch in a supramolecular way to form nanomaterials (Raveendran et al. 2003). Nanomaterials can be incorporated in biopolymers by both sorption and impregnation (Shankar and Rhim 2018). Polymeric nanomaterials are solid colloidal particles within the size range of 10 nm–1 μ m.

Physical properties of nanomaterials can be drastically different from the corresponding macro-sized, bulk material because nanomaterials have much higher surface area and reactivity (Sreedharan and Rao 2019). Either nanospheres or nanocapsules can be prepared, depending on the preparation method (Sharma 2019). Biopolymeric nanomaterials are characterised by microscopy, spectroscopy and other techniques (Fig. 2). A list of green polymeric nanomaterials used for textile dye degradation is presented in Table 1.

Photocatalytic degradation of dyes by biopolymeric nanomaterials

Biopolymers such chitosan act as support material of metallic photocatalysts. Owing to strong adsorption and high surface area, chitosan reduces the amount of intermediates during photocatalytic reactions. In addition, chitosan allows quick and trouble-free recovery of the photocatalyst, which can be recycled with or without any regeneration (Adnan et al. 2020). Photocatalysis is different versus general catalysis in a way that during photocatalysis photons induce catalysis at the time of reaction (Bahal et al. 2019). In the presence of photon (λ), oxygen acts as an electron acceptor and electrons are generated photocatalytically by the breakage of complex dyes (Yang et al. 2005). In response to visible light, polymeric nanomaterials have been shown to degrade

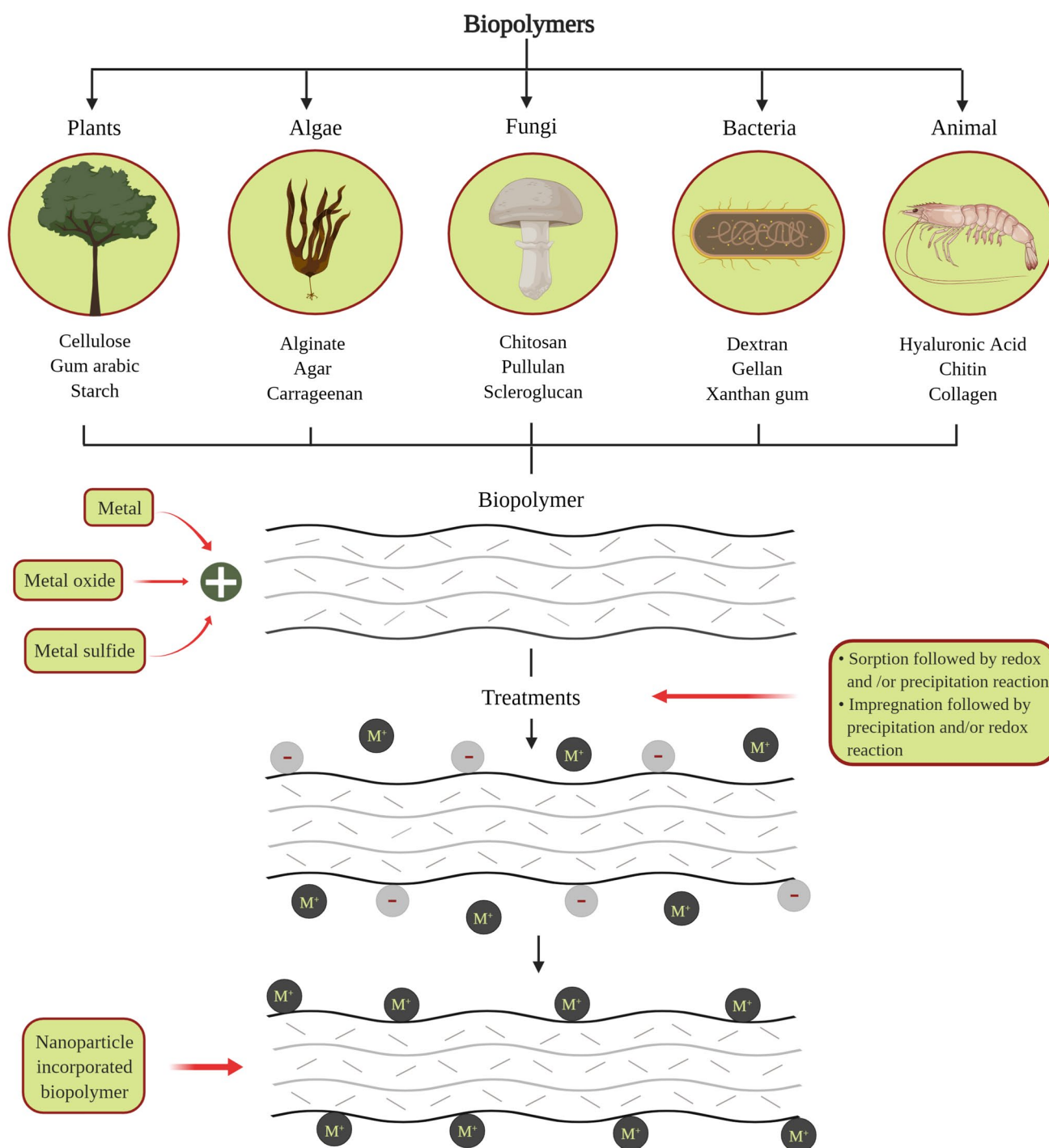


Fig. 1 Different sources of biopolymers and their usage in the synthesis of green polymeric nanomaterials. M^+ : metal ions

dye-containing wastewater photocatalytically within very short period (Bahal et al. 2019), which is both eco-friendly and inexpensive.

The concept of using TiO_2 nanoparticles for photocatalytic dye degradation was developed several decades ago (Fujishima and Honda 1972). Green-synthesised silver nanomaterials have also been used as photocatalysts to treat dyes

and other organic chemicals (Sharma et al. 2009). At that time, nanotechnology was not popular for wastewater remediation, but now it is due to evidence of high performances (Durgalakshmi et al. 2019).

Electron affinity is a major parameter for photocatalytic degradation of reactive textile dyes (Saravanan et al. 2013) as the ionic nature or the presence of lone pair electrons in

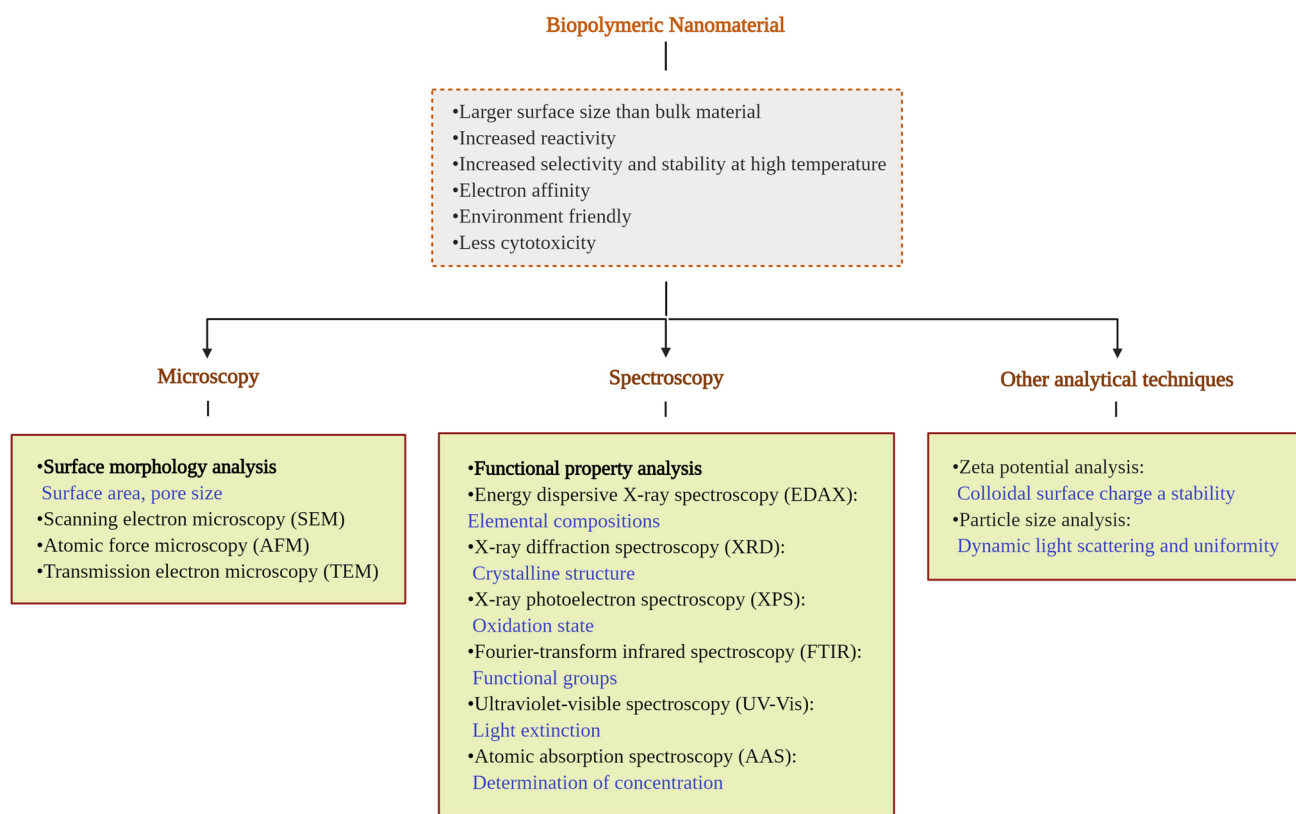


Fig. 2 Properties and analysis of biopolymeric nanomaterials

the polymeric chain backbone acts as chelating agent to stabilise the synthesised nanoparticles (Ng et al. 2013). Additionally, polymeric membrane-incorporated metal nanomaterials have increased hydrophilicity, selectivity, strength and stability at high temperature, up to 200 °C (Ng et al. 2013). Metal oxide nanomaterials incorporating gallic acid are also used in photocatalytic degradation of reactive azo dyes (Sreedharan and Rao 2019).

Metal-incorporated biopolymeric nanomaterials

The synthesis of metal-incorporated biopolymeric nanomaterials is outlined in Fig. 3. Metal-incorporated nanomaterials display high efficacy for the photocatalytic degradation of azo dyes. This can be attributed to their pore size, chemistry of surface plane and ideal mechanical rigidity (Opoku et al. 2017). The basics of photocatalytic degradation involve an electron transfer process coupled with a redox reaction. If the semi-conductivity is modified with metal-incorporated nanoparticles, then the system endorses the charge transport at interface and, in turn, decreases the oxidation of the metal (Subramanian et al. 2001). This process increases the lifetime of electron followed by the augmentation of the reactivity. Surface plasmon resonance (SPR) increases the co-existence of electrons due to the small particle size (Sankar et al.

2015). Polymeric nanomaterials with metal incorporation act as a stabilizer for itself. Metal nanoparticles incorporated with polymeric materials such as resin have found industrial applications as reaction catalysts (Kralik and Biffis 2001).

Metal nanoparticles can also be grafted on different polymeric materials, which increases compactness and stability (Tamayo et al. 2019) and provides a different functionality than that of the metal monomeric nanomaterials (Van Berkel and Hawker 2010). Usually, metal nanoparticles vary in size, whereas incorporation of polymer makes nanoparticle sizes more homogeneous and renders the material more stable. This has been shown during the integration of bacterial cellulose fibres with gold nanoparticles (AuNP).

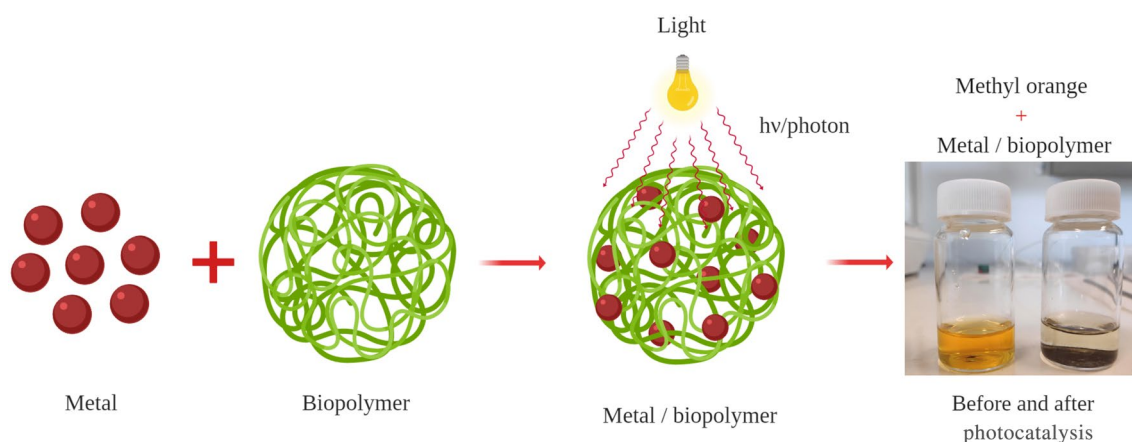
Gold nanoparticles, of 74.32 nm, incorporated in biomaterials from fresh fruiting bodies of the Enoki mushroom degrade nearly 75% of the methylene blue dye in 4 h (Rabeea et al. 2020). Due to their wide substrate specificity, Au nanoparticles are able to treat diverse types of organic dyes (Tamayo et al. 2019). For lanthanum (La), the f-orbital electron of metal ions interacts with different functional groups of different biopolymers and forms complexes with greater surface areas. Lanthanum incorporation in biopolymers results in photocatalytic activity with better adsorption capacity, specifically for organic compounds such as azo dyes (Sirajudheen and Meenakshi 2019). A composite

Table 1 Biopolymeric nanomaterials used for the degradation of textile dyes degradation

Type of nanomaterial	Catalyst	Biopolymer	Degradation of dyes	References
Metal	Silver (Ag)	κ -Carrageenan gum	Mineralisation and catalytic degradation of industrially significant organic dyes such as methylene blue and rhodamine B	Pandey et al. (2020)
	Palladium (Pd)	Chitosan c-Nanotube supported	Congo red, methylene blue, methyl orange, methyl red	Sargin et al. (2020)
	Chitosan/Fe	Chitosan	Basic dye	Kasiri (2019)
	Au	Alginate beads	Discoloration of azo dye acidic orange 7 and reactive orange 5	Ahmed (2019)
	Lanthanum (La)	Chitosan	Photocatalytic degradation of azo dye (methylene blue)	Sirajudheen and Meenakshi (2019)
	Gold (Au)	Bacterial cellulosic fibre	Azo dye degradation	Tamayo et al. (2019), Vilela et al. (2018)
	Copper (Cu)	Chitosan	Congo red	Ali et al. (2018)
	Silver (Ag)	Chitosan	Ponceau BS dye	Sultana et al. (2017)
	Palladium (Pd)	Carboxymethyl cellulose	Degradation of azo dye	Li et al. (2017a)
	Zirconium (Zr)	Gelatine	Methylene blue and fast green	Thakur et al. (2017)
	Palladium (Pd)	Glucuronarabinogalactan polymer and gum olibanum (<i>Boswellia serrata</i>)	Coomassie brilliant blue G-250, rhodamine B, methylene blue and 4-nitrophenol	Kora and Rastogi (2016)
	Copper (Cu)	Chitosan-coated cellulosic microfibres	Methyl orange and congo red	Kamal et al. (2016)
	Metal oxide	ZnO	Chitosan in the form of hydrogel beads	Methylene blue
ZnO		Quince seed mucilage	Photocatalytic degradation of methylene blue	Moghaddas et al. (2020)
Fe ₃ O ₄		Chitosan	Hazardous dye X-3B	Adnan et al. (2020)
MnO ₂		Cellulose	Indigo carmine dye solution	Oliveira et al. (2020)
Alumina (Al ₂ O ₃)		Chitosan	sulfonated azo dye methyl orange	Kasiri (2019)
ZnO		Chitosan	Chromium complex dye, Direct Blue 78, Acid Black 26	Kasiri (2019)
ZnO		Cellulose	Dye-containing wastewater remediation	Bahal et al. (2019)
TiO ₂		Chitosan–acrylic acid biopolymer	Malachite green	Bahal et al. (2019)
ZnO		Cellulose acetate polymeric sheet	Congo red, methyl orange, methylene blue	Khan et al. (2019)
TiO ₂		Bacterial cellulose	Photocatalytic dye degradation	Vilela et al. (2018)
TiO ₂		Cellulose by the fermentation of <i>Komagataei bacterxylinus</i> -immobilized laccase	Reactive red X-3B	Li et al. (2017b)
TiO ₂		Oak gall tannin	Direct yellow 86	Binaeian et al. (2016)
TiO ₂		Chitosan	Acid orange 7, acid red 18, C.I. acid blue 113, reactive yellow 17, reactive black 5, direct blue 78	Škorić et al. (2016)
ZnO	Conducting polyalanine polymer	Methylene blue and malachite green	Riaz et al. (2015)	
Metal sulphide	ZnS	Chitosan	Photodegradation of organic dyes (methyl orange)	Das et al. (2017)
	ZnS	Chitosan	Around 90% photodegradation of methylene blue under UV irradiation	Mansur and Mansur (2015)

Table 1 (continued)

Type of nanomaterial	Catalyst	Biopolymer	Degradation of dyes	References
Others	AgCl	Chitosan in the form of hydrogel beads	Methylene blue	Taghizadeh et al. (2020)
	Fe ₃ O ₄	Immobilised laccase from <i>Bacillus</i> sp. MSK-01 conjugated with thiolated chitosan	Biocatalytic degradation of organic dyes (Reactive Blue 171 and Acid Blue 74)	Ulu et al. (2020)
	TiO ₂	Chitosan–epichlorohydrin	Reactive Red 120	Jawad et al. (2020)
	CuSO ₄	Chitosan-coated nanocomposite from <i>Psidium guajava</i> aqueous leaf extract	Congo red and methylene blue	Sathiyavimal et al. (2020)
	ZnO	Arabic gum-grafted polyacrylamide hydrogel	Complete degradation of malachite green	Mittal et al. (2020)
	ZnO/CuO	Cellulose nanocrystal from bleached bagasse pulp	Rose Bengal (RB)	Elfeky et al. (2020)
	Ag/TiO ₂	Carboxymethyl cellulose and gelatine	Organic dye pollutant	Farshchi et al. (2019)
	SiO ₂	Chitosan/carbon nanotubes	Direct Blue 71, Reactive Blue 19	Kasiri (2019)
	AgNO ₃	Chitosan and guar gum	Binary dye	Vanaamudan et al. (2018)
	ZnS	Chitosan	Photocatalytic degradation of organic dye	Das et al. (2017)
	AgNO ₃	Tangerine peel containing carbohydrate polymers	Catalytic reduction of methyl orange	Alzahrani (2015)
	Pt-TiO ₂	Conjugated polymer	Photocatalytic degradation of azo dye	Dong et al. (2015)

**Fig. 3** Synthesis of metal-incorporated biopolymeric nanoparticles for photocatalytic dye degradation

of lanthanum (La) metal and chitosan has degraded 90% of methylene blue in 40 min (Sirajudheen and Meenakshi 2019). Here, the chemical oxygen demand (COD) decreased nearly 8 times, indicating mineralisation of methylene blue.

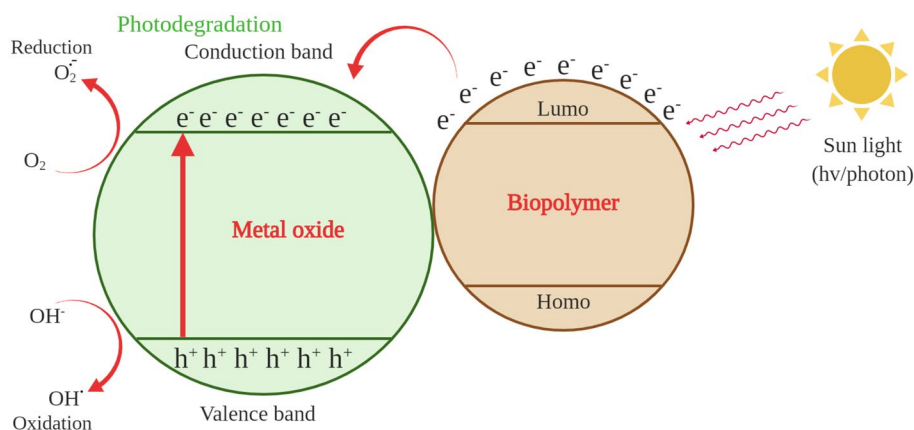
Metal oxide-incorporated biopolymeric nanomaterials

Synthesis of metal oxide-incorporated biopolymeric nanomaterials for photodegradation of dye is illustrated in Fig. 4.

Nanomaterials made up of metal oxide and biopolymers have also extensive photocatalytic activity that can degrade complex chemical structure of azo dyes. Incorporation of biopolymer and metal oxide nanomaterials improves the physicochemical properties of the nanomaterials. Conversely, the presence of metal oxide within the biopolymeric structure enhances the properties of polymer too (Prasanna et al. 2019).

ZnO is one of the most efficient nanomaterials for photocatalytic dye degradation because of ZnO semi-conductivity,

Fig. 4 Synthesis of metal oxide-incorporated biopolymeric nanoparticles for the photodegradation of dyes



stability and activity (Ravishankar et al. 2014). Starch-based ZnO nanomaterial has been reported for its improved conductive and dielectric properties, compared to pure metal oxide nanoparticles (Ravishankar et al. 2014). Cellulose-based ZnO nanomaterials display higher thermal stability than that of the pure metal oxide, and can be used for remediation of dye-containing wastewater at large scale (Ravishankar et al. 2014; Azizi et al. 2013). A chitosan–acrylic acid biopolymer grafted with nano-TiO₂ was reported to degrade more than 90% of malachite green present in wastewater under neutral pH, through a visible light-mediated photocatalytic way (Bahal et al. 2019).

Chitosan incorporating nano-iron oxide (Fe₃O₄) has been used for purification of dye-containing wastewater (Prasanna et al. 2019; Ngah et al. 2011). Chitosan/nano-Fe₃O₄ nanomaterial is also increasing frictions due to magnetic dipole–dipole interactions during the degradation of the hazardous X-3B dye (Adnan et al. 2020). TiO₂ is widely used for preparing biopolymeric nanomaterials due to TiO₂ advantageous surface properties and photocatalysis under visible light (Bahal et al. 2019). Cyclodextrin, an oligosaccharide produced from enzymatic conversion of starch, has been used for wastewater treatment after modification with nano-TiO₂ (Khaoulani et al. 2015). ZnO/carbon black grafted in cellulose acetate has been used to treat azo dyes such as congo red, methyl orange and methylene blue (Khan et al. 2019).

MnO₂/cellulose nanoparticles of size lower than 100 nm degrade 90% indigo carmine within 25 min under ambient light and acidic pH (Oliveira et al. 2020). Here, the biopolymeric nanomaterials can be recovered from solution and recycled for at least 10 times without compromising the decolourisation efficiency. This provides evidence that in the presence of photons, metal oxide-incorporated biopolymeric nanomaterials degrade complex azo dyes within a very short time through an eco-friendly, recyclable process. Few bio-sourced enzymes degrade reactive azo dyes. Laccase shows good potential for bioremediation of

dye-containing wastewater (Wang et al. 2013; Sarkar et al. 2020). Nano-Fe₃O₄/SiO₂ supported with immobilized laccase has achieved nearly complete degradation of the azo dye procion Red MX-5B within 20 min (Wang et al. 2013).

Metal sulphide-incorporated biopolymeric nanomaterials

Synthesis of metal sulphide-incorporated biopolymeric nanoparticles is depicted in Fig. 5. The biopolymer helps to crystallize ZnS nanoparticles (Tiwari and Dhoble 2016). ZnS-incorporated chitosan nanomaterials of 4 nm size display a photocatalytic activity and are used in photodegradation of organic dyes in wastewater (Das et al. 2017). Cadmium (Cd) and lead (Pb) are also used for the formation of metal sulphide nanomaterials. For instance, *Klebsiella pneumonia* has the ability to synthesise electron-dense nano-CdS materials on its cell membrane, which can induce photoreduction of methyl orange by subsequent electron transfer (Das et al. 2017).

Nano-chitosan has been used to remove hazardous dyes (Mansur and Mansur 2015). Chitosan-based quantum dots, a nano-photocatalyst and ZnS are able to remove methylene blue (Mansur and Mansur 2015). Here, ZnS acts as a semiconductor and thus enhances the removal. This ZnS/chitosan-based nanomaterial of nearly 3.5 nm size induces 90% photodegradation of methylene blue by oxidation under UV irradiation within 90 min.

Other biopolymeric nanomaterials

Metal/metal oxide nanoparticles are of special interest because the metal centre of the metal–metal oxide nanomaterial increases the semi-conductivity and rate of separation of electron holes, which in turn increases the photon irradiation, followed by escalation of the photocatalytic activity (Malagutti et al. 2009). For example, 0.25% Ag–TiO₂ thin film incorporation in a resin biopolymer drastically increases

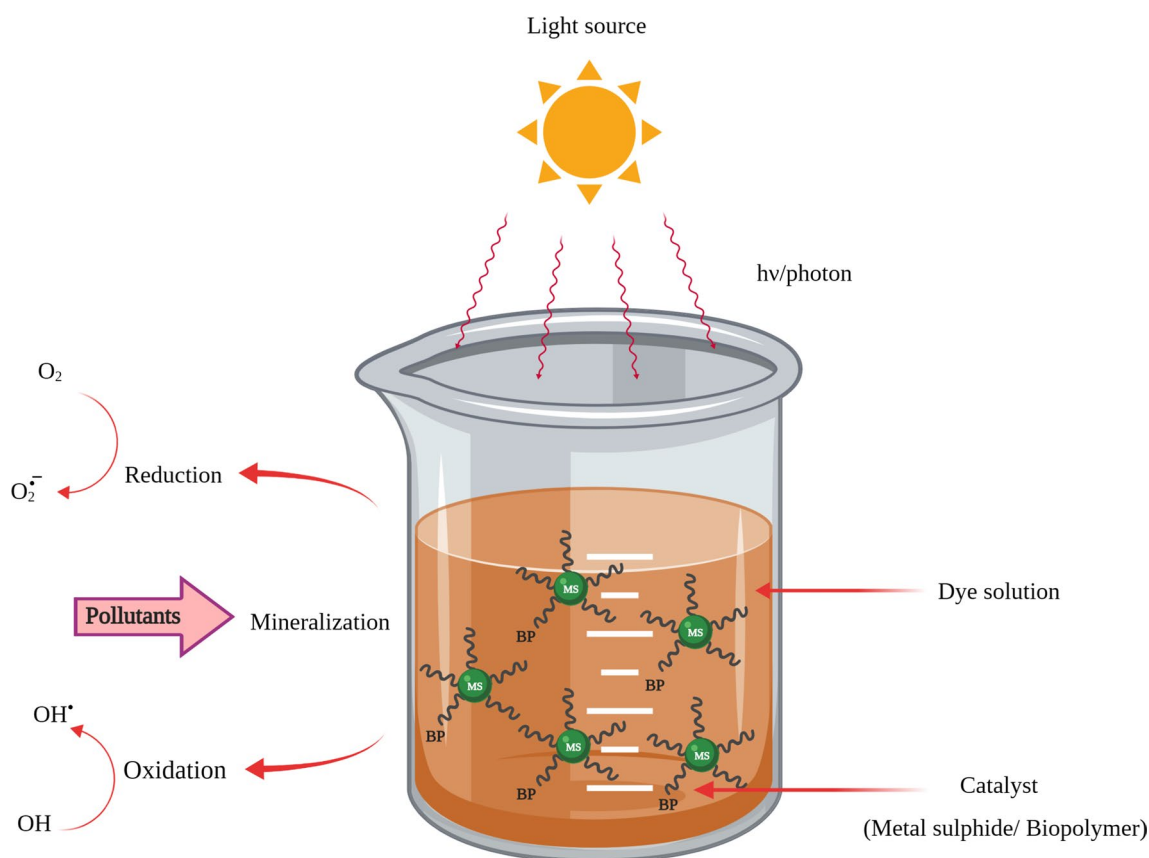


Fig. 5 Rationale behind the synthesis of metal sulphide-incorporated biopolymeric nanoparticles as photocatalysts for dye degradation. BP: biopolymer, MS: metal sulphide

the photocatalytic activity of rhodamine B, compared to sole TiO_2 , as a result of reduced electron hole recombination (Malagutti et al. 2009). TiO_2/Ag hybrid, modified by the incorporation of carboxymethyl cellulose and gelatine of 50–100 nm size, has shown improved photocatalytic activity towards benzene and NH_3 present in the chemical structure of organic pollutants (Farshchi et al. 2019).

Various other chemicals are integrated with diverse polymers for the synthesis of photocatalytic nanomaterials. Several coupled systems expand the photocatalytic performances, such as metal-incorporated metal oxide and dual metal oxide systems (Gnanasekaran et al. 2017, 2018; Sulaiman et al. 2018; Kuo et al. 2019). These systems have two different band positions under sunlight or UV light exposure, which avoids electrons–holes recombination, and thus improves the photocatalytic activity (Gnanasekaran et al. 2017, 2018; Sulaiman et al. 2018; Kuo et al. 2019).

Unique properties of nanocomposites have created a revolution in the field of bioremediation (Mohanraj et al. 2020). Recently, several research groups have tried diverse biopolymeric nanocomposites such as metal/metal oxide/biopolymer or metal oxide/conducting polymer for improved degradation of synthetic azo dyes. One example

of biopolymer/metal oxide nanocomposite is chitosan/ $ZnO/AgCl$ nanocomposite based on hydrogel beads, which permits complete photocatalytic degradation of methylene blue (Taghizadeh et al. 2020). The presence of chitosan in nanocomposites or other biopolymeric nanomaterials significantly increases the degradation activity, as a consequence of hydrophilic adsorption of organic pollutants (Adnan et al. 2020). Furthermore, chitosan-coated $CuSO_4$ nanocomposite, synthesised from *Psidium guajava* aqueous leaf extract, induced more than 90% oxidative photodegradation of congo red and methylene blue within 150 min (Sathiyavimal et al. 2020). Here, electrons are generated from the valance bond due to the presence of sunlight.

Cellulose nanocrystals have been prepared from bleached bagasse pulp and reacted with ZnO/CuO to synthesise biopolymeric dual metal oxide nanocomposites, which can degrade rose bengal more than 99% in 40 min (Elfeky et al. 2020). Immobilised dye-degrading enzyme laccase conjugated with thiolated chitosan– Fe_3O_4 hybrid has been reported to have magnetic properties, and it can remove more than 80% of reactive blue 171 and acid blue 74 within short periods of time (Ulu et al. 2020).

Conclusion

The long-term fate of textile dyes in river and sea sediments is not clear. A recent report suggests that aromatic amines could be naturally degraded in more than 2 years by the sediment bacterial community (Ito et al. 2016). Yet this process is slow and most probably never complete in natural anaerobic environments. Therefore, wastewater should be treated to remove all pollutants before discharge of residual waters in rivers. Here, promising techniques should be based on bio-sourced tools such as microorganisms and dye-degrading enzymes, e.g. laccase, azoreductase or peroxidase (Sarkar et al. 2020). The current success of photocatalytic dye degradation using biopolymers is attributed to the green process, uniform deposition of the nanoparticles, less cytotoxicity and recyclable nature.

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