



## Review Article

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# Bio-derived metal and metal oxide incorporated biopolymer nanocomposites for dye degradation applications: A review

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## ABSTRACT

Water is an essential requirement for both domestic and industrial purposes. The depletion of accessible resources and an upsurge in the water pollution affect both. One of the main causes of water pollution is industrial discharges, specifically the effluents that emanate from textile factories. From these factories, toxic pollutants are often discharged without any monitoring of pollutant control or eradication. The use of the photodegradation technique has successfully demonstrated the efficient decomposition of these toxic pollutants. Different sizes and shapes of materials have already been explored as photocatalysts. Among them, nanometric photocatalysts turned out to be the most effective. To achieve further advances, biopolymers are introduced alongside nanostructured materials for better recycling, biodegradable, and as an eco-friendly approach. This review aims to explore into metal-biopolymer and metal oxide-biopolymer composites from synthesis to application perspectives. We have also elaborated particularly on studies that explore the composition of metals and metal oxides with biopolymers for the application of dye degradation.

**Keywords:** Bio-derived metal and metal oxide; biopolymer; nanocomposites; dye degradation; environmental applications

## 1. INTRODUCTION

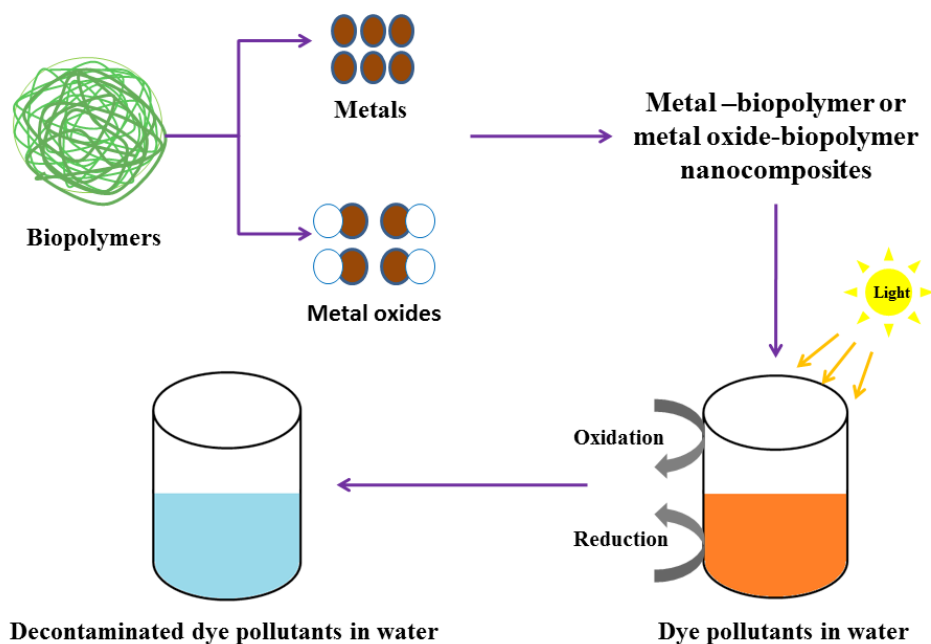
Water is one of the most essential requirements of our daily lives. But water pollution is constantly restricting available consumable sources of water which is creating havoc chaos around the globe. Consumption of water containing hazardous pollutants is equally harmful to humans as well as for animals. Untreated or partially treated water can pose severe health-oriented threats to all living organisms. Edible food staff is also impacted by contaminated water. Ejected contaminants from textile factories are considered as one of the potential threats which can pollute different water sources as the emitted contaminants are mostly emitted in water bodies. These emanations are mainly comprised of synthetic azo dyes which are non-biodegradable, carcinogenic, and recalcitrant by nature (Singh et al., 2015). So, treating them is a crucial concern to stop their further spreading in different water sources and to achieve that various membrane technologies, adsorbents, and photocatalyst materials are already been developed and their advancements are ongoing (Crini and Lichtfouse, 2019; Dutta et al., 2020). Mostly process such as electrochemical oxidation, photocatalysis, ozonation, ion exchange, filtration, and osmosis are considered as

the effective ones to eliminate dye contaminates from water solutions (Singh et al., 2015; Lade et al., 2015).

Photoactivated degradation is a significant technique to eradicate pollutants from water. This process involves the utilization of photocatalysts and light sources. Generation of charge pairs by exposing photocatalyst material to light helps to decontaminate the water solutions *via* oxidation and reduction reactions. The material bandgap is an important factor to determine the working ability of the photocatalysts as photocatalytic degradation involves the transition of electrons between conduction and valence bands. In recent trends use of heterogeneous photocatalysts became an attractive choice for performing dye degradation activity in aqueous or watery solutions. Various oxide materials such as ZnO, CaO, TiO<sub>2</sub>, etc., and their composites are already been explored as promising photocatalyst materials which have exhibited outstanding dye decomposing capability (Fujishima and Honda, 1972; McLaren et al., 2009; Xu et al., 2019; Jaiswal et al., 2021a, 2021b). By coupling or surface modification *via* doping also resulted in enhanced photocatalytic performance (Gnanasekaran et al., 2017; Sulaiman et al., 2018). The addition of natural polymers such as cellulose, starch to metal or metal oxide

photocatalysts have also shown some positive outcomes. Already studies have depicted that natural polymer individually offers efficient adsorption of pollutants (Anastopoulos et al., 2017). For example, chitosan is been utilized for biosorption of toxic dye (Crini et al., 2019). So to further up-gradation of photodegradation activity researchers have utilized the composites by combining metal or metal oxide with biopolymer (Oliveira et al., 2020; Jaiswal et al., 2018; Haldorai and Shim, 2013). However, it can use to be mentioned that several microalgae have been applied to

remove the toxic heavy metals and other pollutants from the contaminated wastewater, also it has been reported for dye removal (Fatima et al., 2020a, 2020b; Jaiswal and Prasath, 2016). Moreover, in few studies photocatalyst materials are synthesized using natural polymers (Sriramulu and Sumathi, 2017; Rabeea et al., 2020). In this review, we are going to elaborate on recent studies on different biopolymer-supported photocatalysts and their workability in degrading dye effluents.



**Figure 1.** Schematic representation of dye degradation applications of metal and metal oxide-incorporated biopolymer nanocomposites

## 2. BIO-SYNTHESIS OF METAL AND METAL OXIDE NANOPARTICLES

Frequently used biopolymers for nanomaterials or composite synthesis are cellulose, chitosan, dextran, and starch (Banerjee and Bandopadhyay, 2016; Farshchi et al., 2019; Kolangare et al., 2019). In sources like bacteria, fungi, algae, and plants, these biopolymers can be found. Ex-situ and in situ techniques are mostly used in synthesizing metal/polymer nanocomposites. Ex-situ involves individual preparation of polymer and metal nanoparticles and afterward combining them *via* employing solution blending method or melt compounding method. On the other hand in situ technique comprises a dispersion of metal nanoparticles in monomeric solution and polymerizing them later on (Tamayo et al., 2019). Another way of in situ preparation is metal ion reduction and thereafter polymerization. To synthesize metal oxide nanoparticles methods such as thermal evaporation, pulsed laser deposition, sputtering

deposition, molecular beam epitaxy, sol-gel, solvothermal etc are used. And to prepare further metal oxide/biopolymer composite methods such as in-situ polymerization, sol-gel approach and direct blending/mixing are utilized (Prasanna et al., 2019). Among these techniques, sol-gel has fewer advantages over other techniques which are high purity, less sintering temperature, and uniform distribution of particles. However, all three processes are quite frequently employed for preparation. In preparation of these composites, few reaction parameters such as temperature, type and amount of solvent, etc are precisely monitored as these impact the morphology of the composite. Apart from conventional techniques, the green synthesis approach are also been mentioned by researchers to avoid the usage of toxic chemical compounds for synthesis purposes (Edison and Sethuraman, 2012).

## 3. METAL AND METAL OXIDE-INCORPORATED BIOPOLYMER NANOCOMPOSITES

Owing to suitable surface chemistry, pore size, and mechanical properties, metal-incorporated nanostructured materials have exhibited excellent photo-degradation of harmful

dyes (Opoku et al., 2017). Incorporating metal into different types of polymeric nanomaterials has elevated transfer of generated charges and hence metal oxidation has been reduced

(Subramanian et al., 2001). By this method electron lifetime as well as reactivity has been increased. Polymeric materials like resin, with metal incorporation, have already been commercially utilized as a reaction catalyst (Kralik and Biffis, 2001). Grafting metal nanomaterials on various polymeric substances can potentially improve stability and compactness (Tamayo et al., 2019). Moreover, the resultant metal incorporated polymeric materials offer better homogeneous size distribution compared to the individual metal nanoparticles. As a result, much stable performance can be achieved. For the first time, Rabeea et al., (2020) used the Enoki mushroom fruit-body extract to prepare extracellular mycosynthesized triangular and spherical gold nanoparticles (Rabeea et al., 2020). The as-synthesized nanoparticles were employed to decompose methylene blue dye. Tamayo and his group mentioned in their studies that gold nanostructured particles have excellent capability of decomposing harmful organic dyes because of having wide substrate specificity (Tamayo et al., 2019). Previously, extracts of the different plants and parts of the plants such as *Pleurotus sajor caju*, *Solanum tuberosum*, *Pleurotus ostreatus*, *T. chebula*, *Agaricus bisporus*, and *Ganoderma lucidum* were used to synthesize different nanostructured materials including silver nanoparticles for investigating the degradation of various dyes and several other applications (Sriramulu and Sumathi, 2017; Edison and Sethuraman, 2012; Nithya and Rangunathan, 2011; Roy et al., 2015; Verma et al., 2021; Ahmad et al., 2020, 2021a, 2021b). Apart from gold and silver, lanthanum was also employed because of its ability to interact with biopolymer-containing functional groups to construct complexes with superior surface area. Sirajudheen and Meenakshi, (2019) have examined the chitosan/lanthanum ( $\text{La}^{3+}$ )/graphite composite to study the decontamination of the methylene blue dye solution (Sirajudheen and Meenakshi, 2019). Similarly, Kusriani et al., (2018) also studied the adsorption impact of lanthanum/chitosan/modified graphite waste composite on various dyes such as methylene blue, rhodamine B, methyl violet, and methyl orange (Kusriani et al., 2018). The author particularly mentioned that a higher concentration of lanthanum (0.03 M) resulted in higher adsorption. The graphene and graphene-based composites have been reported to be easily synthesized using different green route approaches including microwave irradiation for numerous applications (Jaiswal et al., 2018; Sudhakar et al., 2017, 2018).

Besides incorporating metals, incorporating metal oxide with biopolymers has also been proven as an efficient photocatalyst. The combination of biopolymers and metal oxide enhanced physicochemical properties of the resultant nanomaterials which further led to the better decomposing complex chemical structure of toxic dyes and hence superior photocatalytic performance has been achieved (Prasanna et al., 2019). The schematic representation of dye degradation applications of metal and metal oxide-incorporated biopolymer nanocomposites has been shown in Figure 1. ZnO is known for its excellent ability as an efficacious catalyst (Dutta et al., 2019). As a photocatalyst also it has exhibited some noticeable outcomes (Ravishankar et al., 2014). Farzana and Meenakshi, (2015) have examined the degradation

efficiency of ZnO incorporated chitosan beads using Reactive Red 2 dyes (Farzana and Meenakshi, 2015). The author investigated both the individual ZnO and ZnO/chitosan beads under both UV and visible light irradiation. The observation which is needed to be mentioned is that the ZnO under UV radiation and ZnO/chitosan beads under visible light exhibited superior performances. However, the overall highest degradation efficiency of Reactive Red 2 dye was achieved using ZnO catalyst under UV irradiation. Similarly, Haldorai and Shim, (2013) synthesized chitosan encapsulated ZnO to investigate the degradation performance of methylene blue dye (Haldorai and Shim, 2013). Here, the author recycled the ZnO/chitosan hybrid catalyst 5 times to study the impact on the photocatalytic performance of the dye but no specific change was observed. In another study, Khan et al., (2019) investigated the degradation impact of CuAg/ZnO/carbon black/cellulose acetate hybrid nanocomposite on various harmful dyes such as methylene blue, methyl orange, rhodamine B, and congo red out of which congo red have shown the fastest decomposing rate (Khan et al., 2019). Recently Kumar et al., (2021) reported the synthesis of ZnO/SnO<sub>x</sub> photocatalyst and its utilization in the degradation of Methylene blue (Kumar et al., 2021). The author illustrated that tin oxide (SnO<sub>x</sub>) as a coating substance helps to enhance the chemical stability and photo-activity of the composite. The main aim of the study was to prepare ZnO/SnO<sub>x</sub> incorporated in a chitosan matrix, however, the author only employed ZnO/SnO<sub>x</sub> composite to study the degradation and ZnO/SnO<sub>x</sub>/chitosan nanocomposites to study antifouling application. The author further added that ZnO/SnO<sub>x</sub>/chitosan nanocomposites are expected to be exhibiting lesser degradation efficiency compared to ZnO/SnO<sub>x</sub> composite due to the loss of surface area (Kumar et al., 2021).

Apart from ZnO/biopolymer composites, TiO<sub>2</sub> incorporated biopolymer has also indicated workable dye degradation outcomes. Dassanayake et al., (2018) reported the synthesis of aerochitin/TiO<sub>2</sub> and its utilization in the degradation of methylene blue dye (Dassanayake et al., 2018). Aerochitin is an excellent sorbent and that is why incorporating TiO<sub>2</sub> in it resulted in better photocatalytic activity. TiO<sub>2</sub>/chitosan composites are also explored by researchers. Combining chitosan with TiO<sub>2</sub> has resulted in higher catalytic activity and easy recovery. Farzana and Meenakshi, (2014) prepared TiO<sub>2</sub>/chitosan composite to study the degradation of three different dyes (rhodamine B, methylene blue, and reactive red 2). Under UV radiation, reactive red 2 was observed to be almost achieved complete degradation compared to the other two dyes (Farzana and Meenakshi, 2014). To achieve further improvement in surface properties, Bahal et al., (2019) synthesized TiO<sub>2</sub>/chitosan/acrylic acid and employed it to decompose malachite green dye under visible light irradiation (Bahal et al. 2019). The addition of chitosan provided a strong polymeric backbone which helped the functional groups to degrade the dye efficiently. Other than aerochitin and chitosan, TiO<sub>2</sub>/cellulose composites were also recommended by researchers to enhance photodegradation ability (Virukutyte et al., 2012; Han et al., 2017; Uddin et al., 2007; Luo and Huang, 2014; Liu et al., 2010; Khaoulani et al., 2015). For an instance, Han et al., (2017)

prepared TiO<sub>2</sub>/hydroxyethyl cellulose/carboxymethyl hydrogel to examine the degradation of methylene blue dye (Han et al., 2017). The as-synthesized composite not only improved the contact area but also enhanced the interaction probability between the catalyst and the dye. Apart from enhanced photocatalytic performance, the composite also exhibited regenerative capability and self-cleaning properties. Besides ZnO and TiO<sub>2</sub>, other metal oxides such as manganese dioxide, iron oxide, and silver oxide incorporated bio-

composites were also explored for water decontamination application but still requires further investigation with an extensive approach (Prasanna et al., 2019; Ngah et al., 2011; Adnan et al., 2020; Oliveira et al., 2020; Jusoh et al., 2017). Hence the addition of biopolymers into metal or metal oxide facilitated degradation of pollutants as well as offers recyclability and an environmentally safe approach.

**Table 1.** Metal and metal oxide-incorporated biopolymer nanocomposites for dye degradation applications.

Catalysts	Light irradiation	Concentration (mg/L)	Type of dye	Efficiency (%)	Time (min)	References
Bio-Ag nanoparticles	UV	10	direct blue 71	98	150	Sriramulu and Sumathi, 2017
Bio-Au nanoparticles	--	5	methylene blue	75	240	Rabeea et al., 2020
MnO <sub>2</sub> /cellulose	fluorescent	22	indigo carmine	90	25	Oliveira et al., 2020
Chitosan/lanthanide/graphite	UV	30	methylene blue	94	40	Sirajudheen and Meenakshi, 2019
Chitosan/PANI/Co <sub>3</sub> O <sub>4</sub>	UV	10	methylene blue	88	180	Shahabuddin et al., 2015
AgO/chitosan	UV	35	methylene blue	72	180	Jusoh et al., 2017
Chitosan/ZnO	UV	30	methylene blue	64	180	Haldorai and Shim, 2013
Chitosan/lanthanum/graphite	UV	100	methyl violet	85	---	Kusrini et al., 2018
Chitosan/ZnO-SnO <sub>x</sub> /GA	visible	10 <sup>-3</sup>	methylene blue	98	480	Kumar et al., 2021
CuAg/ZnO/carbonblack/cellulose acetate	UV/visible	5×10 <sup>3</sup>	congo red	100	~20	Khan et al., 2019
CuAg/ZnO/carbonblack/cellulose acetate	UV/visible	5×10 <sup>3</sup>	methylene blue	100	~12	Khan et al., 2019
CuAg/ZnO/carbonblack/cellulose acetate	UV/visible	5×10 <sup>3</sup>	rhodamine B	97	~14	Khan et al., 2019
CuAg/ZnO/carbonblack/cellulose acetate	UV/visible	5×10 <sup>3</sup>	methyl orange	100	~12	Khan et al., 2019
Chitosan/polyacrylic acid/ TiO <sub>2</sub>	visible	100	methylene blue	92	240	Bahal et al., 2019
TiO <sub>2</sub> /microcrystalline cellulose	visible	3×10 <sup>3</sup>	methylene blue	90	240	Virkutyte et al., 2012
Hydroxyethyl cellulose/TiO <sub>2</sub> /carboxymethyl cellulose	UV	100	methylene blue	82	30	Han et al., 2017
Aerochitin/TiO <sub>2</sub>	UV	10	methylene blue	98	200	Dassanayake et al., 2018

#### 4. CONCLUSIONS

Degradation of carcinogenic dyes by utilizing photocatalysts is a suitable and precise technique. Here in this review, we have illustrated specifically metal and metal oxide incorporated biopolymers and their work efficiency in degrading the dyes. In actual biopolymers such as chitosan, cellulose, etc., offers a strong backbone to the metal/biopolymer of metal oxide/biopolymer hybrid composites, due to which functional groups can easily facilitate the decomposition of the pollutants. In previous sections, we have explained the frequently used synthesis route and application of various biopolymers composite with metal and metal oxides. Also, details have been explained about

the impact of combining biopolymers to metal and metal oxides on the degradation efficiency compared to their counterparts. In addition to improved photocatalytic activity, the described composite also provided easy recovery and recyclability of the catalysts which endorses commercial usage of these photocatalysts. Commercial usage of these photocatalysts will not only help to treat the dye effluent discharge from the textile industry but also help to clean up other types of harmful pollutants (arsenic, chromium, etc.) usually present in closed water bodies. In this way, we can eliminate threats from water bodies as well as from the environment and be able to move to a safer future.

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## DECLARATION

The authors declare no conflict of interest.

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