



Biopolymer-supported TiO₂ as a sustainable photocatalyst for wastewater treatment: a review

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Abstract

The rising water pollution by pesticides, pharmaceuticals and dyes is a major health issue calling for advanced remediation methods such as photocatalysis with titanium dioxide (TiO₂), yet the use of TiO₂ displays issues of aggregation, mass loss, recovery, and reusability. These issues have been recently solved by synthesizing biopolymer-supported photocatalysts using cheap, biodegradable and safe biopolymers such as chitosan, alginate, cellulose, cyclodextrin, guar gum and starch. Here we review biopolymer-supported TiO₂ photocatalysts for the removal of organic compounds, with focus on preparation methods, photo and chemical stability, reusability, and adsorptive capacity. We discuss applications of immobilized photocatalysts at the industrial scale.

Keywords Alginate · Bio-polymer · Chitosan · TiO₂ · Wastewater

Introduction

Water is a necessity for all humans living on the globe. Freshwater availability has been seriously reduced in recent decades as a result of population increase, industrialization, and growing demand (Gusain et al. 2019). Water contamination is the most serious threat to the growth and survival of flora and animals, causing enormous socioeconomic difficulties (Ponnuchamy et al. 2021). Water contamination increased owing to the presence of both man-made and natural pollutants in water resources. The organic pollutants (pesticides, pharmaceuticals, chlorinated solvents, dyes), inorganic pollutants (nitrates, phosphate, and heavy metals),

and pathogens (coliform bacteria) have been identified as major pollutants in water (Dong et al. 2020; Laws 2017; Yang et al. 2017). Due to the rapid usage of these products, organic pollutants are detected in the different environmental spheres (Dhaka et al. 2019; Jaafarzadeh et al. 2017). These organic pollutants persist in nature, resist biodegradation, and eventually accumulate in the food chain (Gui et al. 2014; Qiu et al. 2019). Therefore, the removal of organic pollutants and their metabolites from water resources are necessary to provide safe drinking water and ensure safety.

The ultimate requirement for adequate water treatment technology is critical for the mineralization of organic pollutants. In recent times, advanced oxidation processes have emerged as powerful technologies in mineralizing the organic contaminants in liquid streams. Among different photocatalysts, TiO₂ is the most widely employed photocatalyst because of non-toxicity, greater photostability, chemical inertness, and better oxidation efficiency (Kanan et al. 2020). TiO₂ is capable of eliminating both organic and inorganic pollutants but they are not commercialized due to different technical barriers. Firstly, the wide band gap (3.2 eV for anatase phase) of the TiO₂ needs ultraviolet light for the activation of photocatalyst and results in minimum utilization of solar light. In the solar spectrum, only 5% of the sun's energy belongs to the UV range, while it is 45% for the visible range (Balakrishnan et al. 2021). Thus, shifting the bandgap of the TiO₂ from the ultraviolet region to the visible region is necessary for real-life applications (Dong

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et al. 2015). Other associated problems with TiO₂ based photocatalysis are (i) low adsorption of organic pollutants, (ii) difficulty in the recovery of photocatalyst, (iii) aggregation of nano-sized catalysts and, (iv) difficulty in uniform distribution of catalyst (Balakrishnan et al. 2020a). The development of new photocatalysts is evident to extend the absorption wavelength to the visible range region and minimize the effects of recombination of electron/hole pairs. In addition, a special focus must be given to the reusability and recoverability of the photocatalyst.

Different strategies have been proposed to overcome the drawbacks of TiO₂ are doping with metals and non-metals (Juan et al. 2021), co-doping (Al-Mamun et al. 2019), heterojunction construction (He et al. 2021), surface organic modifications (Park et al. 2013), and immobilization of support structures (Shi et al. 2019). Where immobilization of support structures is considered as an effective strategy to minimize the cost associated with recovery and separation of photocatalyst from aqueous solution (Sraw et al. 2018). Many studies reported the utilization of different supports (polymers, clay, silica, and perlite) for the photocatalyst to enhance recovery, reusability, and degradation efficiency to a greater extent (Sraw et al. 2018). One of the prominent methods to enhance the reuse of the photocatalyst is the utilization of biopolymers as a support material for TiO₂. The key features of biopolymers include low-cost, durability, environmentally friendly, higher adsorption property, availability and biodegradable (Balakrishnan and Chinthala 2022). This paper reviews the different biopolymers and their performance as support materials for TiO₂ based photocatalyst. The important biopolymers reviewed in this paper are chitosan, cellulose, alginate, cyclodextrin, gum, starch and collagen.

The manuscript is divided into different sections. Firstly, the photocatalytic degradation mechanism of TiO₂ based materials is described. The next sections discuss the photocatalytic supports with potent advantages and disadvantages. The succeeding section briefs the role of biopolymer as a support material for TiO₂. Finally, a comprehensive review is conducted on the removal of organic pollutants using different biopolymer supported photocatalysts. An approach for the scale-up of photocatalytic studies from batch mode to continuous mode is illustrated in the section industrial perspectives. The strategies to overcome the challenges faced by biopolymer supported TiO₂ is also emphasized in the conclusion. It is also believed that this review helps the researchers to scale up the photocatalytic technology.

Photocatalytic degradation mechanism

Heterogeneous photocatalysis is conducted in five stages:

1. Diffusion of organic pollutants from the bulk liquid phase to the photocatalyst surface,
2. Adsorption of organic pollutants onto the surface of photocatalyst,
3. Photochemical reaction due to the interaction between light and catalyst producing highly reactive species (for example oxygen radicals),
4. Desorption of products from the surface of photocatalyst,
5. Removal of products from the bulk of the solution (Paumo et al. 2020).

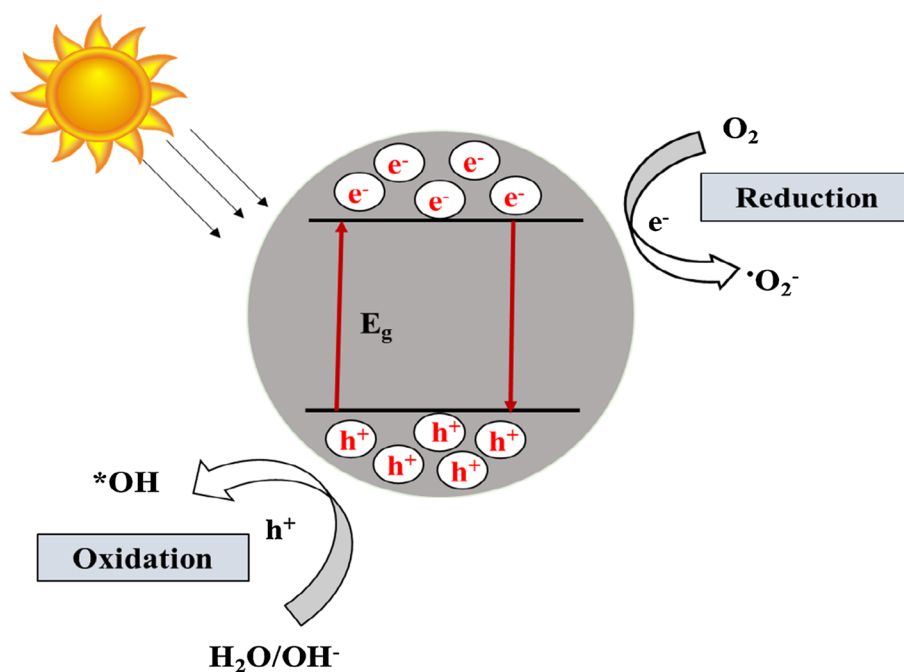
Among these five steps, the slowest step determines the reaction rate for any process. So, the mass transfer steps I and V are speeded than the reaction steps 2, 3, and 5. Steps 2 and 3 do not interfere with the overall rate of any chemical reaction. In photocatalytic remediation, highly powerful hydroxyl radicals are involved as the oxidizing agents for removing pollutants (Al-Mamun et al. 2019).

Upon the illumination by a suitable light source, the electrons are transferred from the valence band to the conduction band with simultaneous generation of holes in the valence band (Fig. 1). These photo-induced holes are easily trapped by hydroxyl ions and are partially converted to hydroxyl radicals quickly (Chinthala et al. 2021). The produced hydroxyl radicals are extremely strong oxidants responsible for the mineralization of organic pollutants (Aanchal and Basu 2020). The highly efficient trapped oxygen may suppress the recombination of photo-induced holes. Moreover, the limited oxygen supply may cause faster recombination of electrons and holes (Dong et al. 2015; Meenakshi & Farzana, 2013; Paumo et al. 2020).

Photocatalyst supports

Photocatalysts take part in a chemical reaction only upon light source irradiation (Shan et al. 2010). Various materials are explored to modify the TiO₂ or other photocatalysts to redeem organic pollutants (Chen et al. 2020a, b). Polymers were incorporated with photocatalysts through immobilization, surface coating, chemical or blending processes (Bet-Moushoul et al. 2016). Any ideal supports for the photocatalyst must exhibit a strong affinity between the photocatalyst and support for anchoring the base material (Singh et al. 2013). The inorganic carbon fabrics, synthetic fabrics, plastics, fly ash, fiberglass, silica gel, wool, glass beads, quartz optical fibers, alumina clays, cellulose, zeolites, chitosan, glass plates, Raschig rings, chitosan, and alginate have been used as support materials for photocatalyst (Adnan et al. 2019; Singh et al. 2013). The different types of support materials and their advantages and disadvantages are mentioned in Table 1.

Fig. 1 Photocatalytic degradation mechanism using TiO₂ as a photocatalyst. The formation of holes in the valence band and the electrons in the conduction band end up in the oxidation of water molecules and consequently, oxygen molecules are reduced. These redox products undergo a reaction with pollutant molecules and give carbon dioxide and water molecules as end products. Where E_g is the band gap of the photocatalyst, h⁺ is hole and e⁻ is electron



Biopolymer as a support material

The development of appropriate and effective ways for hybrid matrices to function as envelopes for dopants of large solid particles (Sirajudheen et al. 2021). Biopolymers, activated carbon, silica, and clays were the most commonly utilized support materials in developing hybrid composite materials (Balakrishnan et al. 2021). The

removal of organic pollutants depends on the form and morphology of the produced photocatalyst. The support materials may also affect the specific surface area of the photocatalyst that could influence the degradation rate of the pollutant.

Apart from improving the specific surface area, biopolymers possess functional groups in their structure that may interact with the organic pollutants and result in their higher removal rate (Nithya et al. 2014; Shukla et al. 2013). The key

Table 1 Types of TiO₂ support materials and their advantages and disadvantages

Support	Example	Advantage	Disadvantage	References
Metal	Copper, platinum, chromium, and gold	Reduce the band gap of the photocatalyst Improve the adsorption of pollutants Minimize electron–hole recombination	Expensive Thermal instability High mass loss and poor reusability	Saqib et al. (2016), Chen et al. (2020a)
Carbon	Activated carbon, graphene, carbon nanotubes, and biochar	High surface area Minimize electron–hole recombination Reduction in band gap Greater adsorption of pollutants	Problems associated with recovery and reusability of catalyst	Bel Hadjltaief et al. (2015), Chen et al. (2020a), Zahedifar and Seyedi (2022)
Ceramic	Silica, zeolite, alumina, and silica carbide	Easy to recover Good photostability Greater oxidation stability	Lower activity of the support than the photocatalyst	Danfá et al. (2021)
Polymer	Chitosan, polylactic acid, polyamide, and polyester	Improve the reusability of the photocatalyst Minimize the mass loss Tune the band gap Prevent aggregation of photocatalyst	Problems due to mass transfer restrictions	Balakrishnan et al. (2020a, b)

reasons for the selection of biopolymers as a suitable support for TiO₂ based materials are (Puls et al. 2010; Singh et al. 2013; Shan et al. 2010):

- Biopolymers are relatively cheap and abundantly available on earth.
- Biopolymers are safe, chemically inert, and mechanically stable with greater durability.
- The majority of the polymers are resistant to ultraviolet and do not participate in the oxidation reaction.
- Bio-polymers as support materials improve adsorption and subsequent oxidation due to the preconcentration of organic contaminants on the catalyst surface.

Biopolymers have gained increased scientific relevance because of their non-toxicity, compatibility, flexibility, pore size, surface morphology, and mechanical strength. As a result, biopolymers are favored over chemical polymers since they are more ecologically friendly and contribute to a more sustainable environment (Karimi-Maleh et al. 2021; Qamar et al. 2020). Due to these unique properties, biopolymers found prominent applications in packaging, biofuels, water treatment, energy storage (George et al. 2020). Majority of the biopolymers are extracted from different biological species as shown in Fig. 2, and Table 2 discusses the properties of the commonly used biopolymer supports.

Preparation methods

The photocatalytic remediation of wastewater has been thoroughly researched by utilizing various biopolymers. The presence of different or unique functional groups indicates the reactivity and flexibility of the polymeric chains for varying applications (Balakrishnan et al. 2020a, b). Different forms of structures can be developed for photocatalytic application. Some of them are beads, films, aerogels, hydrogels, composites, and membranes. Different preparation routes of TiO₂/biopolymer supported photocatalyst are illustrated as follows:

Hydrothermal method

The photocatalyst is synthesized in a steel autoclave at 130 to 250 °C by distinctively crystallizing the substances at an elevated vapor pressure of 0.3 to 4 MPa (da Silva et al. 2020; Denkbaş et al. 2016; Sirajudheen et al. 2021). The crystallization leads to particles agglomeration as the vapors pressure gets saturated. However, the usage of stabilizing agents avoids the problems related to agglomeration (Broujeni et al. 2021; Shao et al. 2019). The hydrothermal method can control both the size and shape of the particles through the use of suitable reagents. Furthermore, this eco-friendly approach requires a one-step preparation route (Jadoun et al. 2021).

Fig. 2 Biopolymers and their sources. The biopolymers are broadly divided on the basis of their sources into animals, higher plants, algae and microorganisms. Based on the applications, animal and plant sources are extensively used for different applications including wastewater treatment




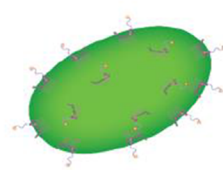
Biopolymers	
<p>Higher Plants: Starch, cellulose, guar gum</p> 	<p>Animals: Chitin and chitosan</p> 
<p>Algae: Alginate and carrageenan</p> 	<p>Microorganism: Xanthan gum, and bacterial cellulose</p> 

Table 2 Commonly used biopolymers for TiO₂ support

Biopolymer	Formula	Functional group	Properties	References
Chitosan	(C ₆ H ₁₁ NO ₄) _n	Hydroxyl group and amino groups	Biocompatible, non-toxic, low cost, and biodegradable	Sirajudheen et al. (2021)
Alginate	(C ₆ H ₈ O ₆) _n	Carboxylate side groups	Biodegradable, biocompatible, and very low immunogenicity	Wang et al. (2019)
Cellulose	(C ₆ H ₁₀ O ₅) _n	Hydroxyl group	Biocompatible, non-toxic, abundant, odorless, insoluble in water, and biodegradable	Hamidon et al. (2022)
Cyclodextrin	(C ₆ H ₁₀ O ₄) _n	Ether group and hydroxyl group	Biocompatible, non-toxic, low cost, and biodegradable. Hydrophobic interior and hydrophilic exterior characteristics	Nasrollahzadeh et al. (2021)
Collagen	C ₅₇ H ₉₁ H ₁₉ O ₁₆	Amino acids	Biocompatibility, biodegradation	Kumar et al. (2020)
Starch	(C ₆ H ₁₀ O ₅) _n	Ether group and hydroxyl groups	Biocompatible, non-toxic, abundant, odorless, insoluble in water, and biodegradable	Kumar et al. (2020)

Sol–gel method

The sol–gel method comprises the condensation of precursors (metal chlorides, nitrates, or alkoxides) in to a solid material upon the evaporation of the respective solutions (Grossin 2021). This is followed by cross-linking reaction and dehydration. Either hydrolysis or condensation of respective precursors yields a sol and coverts to gel upon thermal treatment. Finally, gel is calcined to yield the final products (D'Arienzo et al. 2017). The main advantages of this method are tunable or controllable composition and morphology, low-temperature synthesis, larger surface area, and smaller particle size (D'Arienzo et al. 2017; Yilmaz & Soylak, 2020).

Co-precipitation method

In this strategy, the nanoparticles (metals) are prepared under the presence of biopolymer through precipitation of the catalyst from their precursors (Ashik et al. 2018). The hindered release of anions and cations is beneficial in controlling the nucleation and particle growth. Researchers reported the utilization of this strategy in the preparation of chitosan-based nanocomposites or other structures due to the presence of different functional groups present in their structure (Sirajudheen et al. 2021). The advantages are the ability to control particle size, low-temperature requirements, greater energy efficiency, and the ability to change the surface of particle state (Cruz et al. 2018; Huang et al. 2019). However, this method faces problems due to larger time requirements, and chances of precipitation of impurities present with products (Nawaz et al. 2019).

Immobilization

In this method, the nanoparticles are immobilized onto the surface of polymeric networks. Different strategies used for

immobilizing TiO₂ on biopolymers are crosslinking, entrapment, encapsulation, and covalent bonds (Kumar et al. 2020). All the naturally derived biopolymers can act as supports for immobilizing TiO₂ photocatalyst. The immobilized photocatalyst always displayed phenomenal catalytic stability and high dispersion. This method minimizes the catalyst loss and improves the reusability of the material to a greater extent (Satoh et al. 2016). Even though immobilized photocatalysts are more expensive than homogeneous materials, the cost is reduced via recycling or reusing.

Electrospinning

Electrospinning is a well-adopted technology for preparing fibers or one-dimensional materials through the utilization of polymers and their respective precursors. This method employs the synthesis of a non-woven web of micro or nanofibers (Kumar et al. 2020). High voltage requirement is necessary to apply on the liquid stream and a collector, which gradually allows solution passage through a nozzle. Later, the formed jet-shaped fibers are passed through the drying stage and eventually deposits fibers at the collector (Reddy et al. 2016). The nanofibers prepared through this method have a tunable morphology, porosity, higher surface area to volume ratio, low-cost setup, simplicity, high efficiency. The usage of organic solvents and hindered control of porous structures are drawbacks of the electrospinning synthesis process (Kumar et al. 2020; Sirajudheen et al. 2021).

Phase inversion

The phase inversion is a demixing process, where homogeneous polymer solution is transferred from liquid to solid state in a controlled manner (Zare and Kargari 2018). The conversion is attained through the casting of the polymeric solution on a suitable support, followed by dipping

or coating in a non-aqueous solvent medium (Sirajudheen et al. 2021). Phase inversion is widely used to prepare the polymeric porous membranes with a bulky form of structure (Figoli et al. 2016). The properties and chemical interactions of the synthesized membranes are dependent on the choice of polymers and the type of additives employed in the casting solution (Purkait et al. 2018).

Biopolymer-supported TiO₂ photocatalysts

Chitosan

Chitosan is the second most abundant natural biopolymer in the world (Divya and Jisha 2018). Chitosan is produced from the deacetylation of chitin, which is found in abundance in the exoskeletons of crustaceans (For example shrimp, crab, prawns, and shellfish) (Morin-Crini et al. 2019). The primary benefits of choosing chitosan in wastewater treatment are its low cost and environmental friendliness, high chemical and thermal stability, nontoxicity, biodegradability, simple physical and chemical modification, biocompatibility, and good adsorption properties (Lee et al. 2015). Due to these unique characteristics, chitosan is employed as possible catalyst support. The literature reported that chitosan could be easily modified physically or chemically into different shapes. Because of the hydroxyl and amine groups in the chitosan structure, chitosan is significantly insoluble in certain solvents, making chitosan suitable for the redemption of organic pollutants (Balakrishnan et al. 2020a, b). Additionally, chitosan finds versatile applications in water treatment, biomedical applications, pharmaceuticals, cosmetics, food industry, packaging's and drug delivery (Parhi 2020; Nasrollahzadeh et al. 2021; Sirajudheen et al. 2021).

Additionally, chitosan is used as an adsorbent to remove organic pollutants from aqueous streams. So, incorporating chitosan to TiO₂ integrates both adsorption and photocatalysis. Many studies emphasized the exploration of TiO₂/chitosan-based macroporous materials for different advanced oxidation processes in wastewater technology. Hasmath Farzana and Meenakshi, 2014, described the utilization of chitosan/TiO₂ nano-composites for the destruction of dyes. The Ti–O–C bond enhanced the polarity of the chitosan-TiO₂ mixture benefitting in the efficient separation of electron/hole pairs in the excited state (Hasmath Farzana and Meenakshi 2015). However, a significant catalyst loss was associated with the recovery. Saravanan et al. (2018) emphasized the preparation of TiO₂/chitosan via the sol–gel method at different weight ratios. With increase in the chitosan loading in the nano-composites, a reduction in the bandgap of the photocatalyst was observed. For instance, the amphoteric chitosan/TiO₂ was prepared by grafting the acrylic acid on the chitosan surface under the presence of

potassium persulfate through a free radical polymerization reaction for the elimination of dyes. The obtained chitosan/TiO₂ could remove 99.5% of malachite green within 240 min (15 Watts white light). The higher efficiency of the photocatalyst is attributed to the minimized electron–hole recombination. The functional groups of chitosan also contributed to the degradation of malachite green (Bahal et al. 2019).

For instance, Jbeli et al. (2018) synthesized highly active chitosan-TiO₂/ZnS via immobilization method followed by hydrothermal treatment. Initially, the chitosan was coated over a glass substrate followed by the deposition of titanium precursor and immersed in Zn(O₂CCH₃)₂ and Na₂S. The chitosan, hydroxyl and amino groups and the absence of water molecules accelerated the chemical coordination of titanium precursor, resulting in a small nano-metric amorphous oxo-titanate. Later, Shao et al. (2019) emphasized the development of polydimethylsiloxane-SiO₂-chitosan@TiO₂ composites on pumice through the immobilization method. The polydimethylsiloxane-SiO₂-chitosan@TiO₂ was used to remove methylene blue under the illumination of simulated sunlight. This catalyst also exhibited a reusability of five cycles. Moreover, this immobilization approach has increased photostability, reusability, and minimized filtration costs.

Afzal et al. (2019) focused on the magnetically separable TiO₂/chitosan assisted visible-light mediated photocatalytic degradation of o-nitrophenol. Response surface methodology analysis estimated 92% of nitrophenol removal within 180 min. Moreover, studies proved that the pH of the medium had played an essential role in the elimination of nitrophenol. The ternary chitosan-based catalyst exhibited greater interactions between each component and resulted in higher absorption capacity. However, studies on the reusability of the photocatalysts are not explored. Later, Afzal et al. (2021) highlighted the preparation of TiO₂/chitosan/Fe₃O₄ to eliminate nitrophenol. The studies also proved the reduction in bandgap from 2.85 eV and minimized charge carrier separation. Under the illumination of the solar light, more than 90% of nitrophenol was degraded within 180 min. After four consecutive cycles, the reusability studies showed 1% nitrophenol reduction.

The immobilized TiO₂ on chitosan beads through the phase inversion method was reported by Balakrishnan et al. (2020a, b). With the addition of chitosan to the mixture, the bandgap was reduced from 3.2 to 2.79 eV. Under ultraviolet light, the obtained beads degraded 92% of the 2,4-dichlorophenoxyacetic acid. The studies also answered the concerns of catalyst loss and recovery, as the catalyst demonstrated reusability for ten consecutive cycles with a mass loss of less than 5%, as shown in Fig. 3a. Fourier transform infrared spectra confirmed that the catalyst offered the same stability even after ten cycles (Fig. 3b). They also stated that TiO₂/chitosan beads could be adopted for the continuous mode of

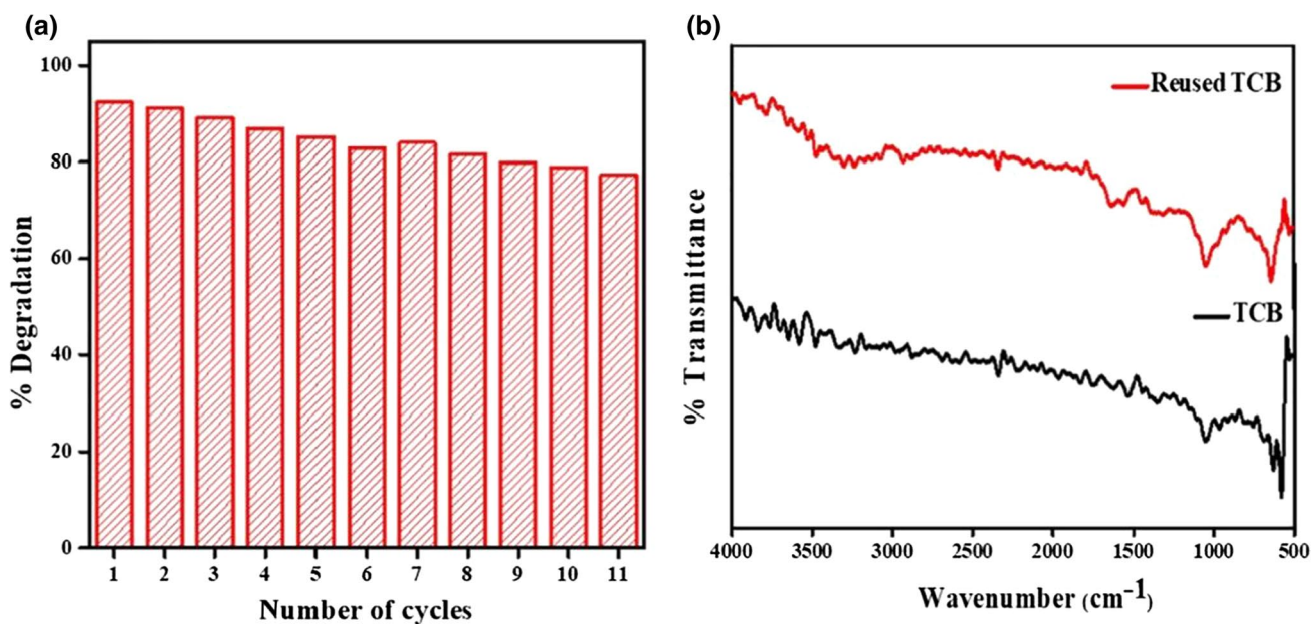


Fig. 3 **a** Reusability of chitosan/TiO₂ beads. A decline in degradation efficiency is observed from 92 to 80% at the end of 11 cycles, **b** FTIR spectra of fresh and reused catalysts confirm the stability of the pho-

tocatalyst. Reprinted from Balakrishnan et al. (2020a, b) with permission from Elsevier. Where TCB is TiO₂/chitosan beads

operation with a potent scope of commercialization. Later, Balakrishnan et al. (2021) also studied the feasibility of 2,4-dichlorophenoxyacetic acid removal (86%) in a circulating type photocatalytic reactor under ultraviolet light. Recently developed TiO₂/chitosan/glycerol beads which removed 93.3% of acid blue 193 at optimized conditions of 90 min, pH 4.5, and an initial concentration of 10 ppm (Hoang et al. 2021).

Xu et al. (2021) reported a simple and highly reusable visible-light-driven chitosan/TiO₂@g-C₃N₄ nanocomposite membrane. The membrane removed methyl orange, rhodamine-B, and chromium (VI) with efficiency greater than 90%. The catalyst also displayed reusability up to 10 consecutive cycles. The membranes hydrophilic nature enhanced the reusability of the photocatalyst without any loss in degradation efficiency. This catalyst was also extended to a continuous flow-photocatalytic system and showed removal of 90% for rhodamine-B. Saleh et al. (2021) developed a bentonite/Chitosan/TiO₂ heterostructure to reduce arsenic (III) present in the water. Upon the illumination of ultraviolet light, the prepared material reduced 97% of arsenic (III) to arsenic (V). The hydroxyl radicals present in the TiO₂ structure were responsible for the pollutant elimination. Moreover, the existence of different co-ions did not interface with the degradation process.

Fused modeling three dimensional deposition, an easily scalable technology is also adopted for the immobilization of TiO₂ nanoparticles. Along with that, three dimensional printing technology also plays a vital role in water

technology. Bergamonti et al. (2019) highlighted the utilization of three dimensionally printed TiO₂/chitosan scaffolds to eliminate amoxicillin under ultraviolet light irradiation. The three dimensionally printed TiO₂/chitosan scaffolds showcased the possibilities to modulate the shape and size of the photocatalyst. This, in turn, increases the surface area to volume ratio, which imparted the photocatalytic degradation of amoxicillin. The literature also proved that the scaffolds are elastic, durable, and robust and can be easily recovered and reused from large quantities of polluted water. Table 3 summarizes the different chitosan-supported TiO₂ photocatalysts for removing organic pollutants from wastewater.

The photocatalytic effectiveness of chitosan-based materials was higher under both visible and ultraviolet light sources. Furthermore, chitosan present in the chitosan-TiO₂ can (i) control the bandgap, (ii) make recovery easier, (iii) decrease mass loss, (iv) enhance stability, and (v) can form any shape or structure (Zhao et al., 2018). Despite the advantages, chitosan possesses several limitations concerning the protonation of NH₂ groups of chitosan. The mutual interaction of the chitosan/TiO₂ photocatalyst through chelation, coordination of NH₂ groups, electrostatic interaction hinders the adsorption of organic pollutants on the catalyst surface. Along with that, most of the studies reported its reusability up to 10 cycles only.

Several steps must be adopted to overcome the problems associated with chitosan/TiO₂ materials in wastewater treatment. Firstly, physical and chemical modification of chitosan is desirable to enhance the physicochemical properties and

Table 3 Chitosan-supported TiO₂ photocatalysts for the removal of pollutants

Catalyst	Pollutant	Light source	Initial concentration (mg/L)	Contact time (min-ute)	Efficiency (%)	References
Chitosan-TiO ₂ composites	Methylene blue	Medium pressure Mercury lamp	50	30	90.9	Hasmath Farzana and Meenakshi (2014)
Silver-TiO ₂ /γ-Al ₂ O ₃ /chitosan nanocomposites	Nitrate	Ultraviolet	100	5	69.6	Zarei et al. (2020)
TiO ₂ /MgO nanocomposites immobilized chitosan hydrogel	Methyl orange, alizarin red S	Ultraviolet	5	90	82.4 41.8	Arikal and Kallingal (2019)
TiO ₂ /chitosan-montmorillonite	Methyl orange	Home fluorescent lamp (45 Watt)	20	60	91.2	Bahrudin and Nawi (2019)
Novel TiO ₂ /chitosan/reduced graphene oxide	Methyl orange	Ultraviolet	250	–	97	Chen et al. (2017a, b)
TiO ₂ /Chitosan	Phenol	Ultraviolet	100	5	33.85	Wardhani et al. (2018)
Nano-TiO ₂ /chitosan/poly(<i>N</i> -isopropylacrylamide) composite hydrogel	Acid fuchsin	Ultraviolet	10	160	90.5	Zhou et al. (2017)
Polyvinyl alcohol/TiO ₂ /chitosan/chlorophyll bi-nano composite film	Methylene blue, 4-chlorophenol, congo red	Light emitting diodes (70 Watt)	10	90	96.2	Soltaninejad and Maleki (2021)
			120	210	94	
			10	120	91	
TiO ₂ coated magnetic polyacrylamide grafted carboxymethylated chitosan	Congo red	Light emitting diodes (12 Watt)	20	180	99.2	Abdelwahab and Helaly (2017)
TiO ₂ -chitosan supported glass plate	Methyl orange	Ultraviolet (6 Watt, 365 nm)	10	60	78	Amir et al. (2017)
Chitosan modified nitrogen, sulfur-doped TiO ₂	Tetracycline	6 Light emitting diodes (18 Watt)	10	20	91	Farhadian et al. (2019)
Chitosan/polyvinyl/sodium-titanate/TiO ₂	Methyl orange, congo red	Ultraviolet	33	15	100	Habiba et al. (2016)
			15	12	99	
Hybrid chitosan-TiO ₂	Aniline	Halogen lamp (47 Watt)	–	540	95	Hamden et al. (2016)

influence the adsorption capacity of the prepared material. Secondly, the doping of metals onto the chitosan/TiO₂ may also improve the activity of the photocatalyst via shifting to the visible light activity, which may also suppress the recombination rate. Finally, the development of magnetic chitosan/TiO₂ materials may also resolve problems related to catalyst recovery.

Alginate

Alginate is an anionic poly-3-saccharide that may be found in the outer shells of certain algae (brown algae, kelps) (Ibrahim et al. 2017). Alginic acid is the main component for alginate, and sodium alginate is the sodium salt of alginate rich in free hydroxyl and carboxyl groups found in the polymer

chain (Wang et al. 2019). The alginate is a copolymer comprised of both α-L-gluronic acid and β-D-mannuronic acid with homopolymeric regions of G block and M-blocks which are connected alternatively with the polymeric structure (Hasnain et al. 2020). Alginate is derived from different sources and exhibits different chemical structures with varying concentrations of G and M-blocks resulting in higher and lower rigidity (Jahandideh et al. 2021).

Sodium alginate is non-toxic and ecologically safe, with high stability due to its strong gelation, film-forming, and complexing properties (Wang et al. 2019). Alginate is biocompatible, biodegradable, non-toxic, and non-immunogenic. Alginate can crosslink with cations due to its intrinsic properties. As a result, alginate is used in various applications in the field of agriculture, food technology, cosmetics,

chemical engineering, environmental engineering, medicine, and others (Subekti and Helmiyati 2018; Szevalska et al. 2016). The environmental remediation employing alginate highlighted the rich functional groups that may improve the uptake of metallic ions through ion exchange between the target pollutant and crosslinking cations. Alginate-based polymers are being used to remove dyes, heavy metals, pesticides, and other emerging contaminants found in water. Due to high rigidity and poor elasticity, surface modifications were carried out through metal doping and organic coatings to overcome the concerns (Wang et al. 2019).

In recent years, different studies explained the usage of alginate as a support for the development of semiconductor-based photocatalyst targeting wastewater treatment (Cheng Keong et al. 2017; Reveendran and Ong 2018). Albarelli et al. (2009) explained the usage of calcium alginate as a supporting substrate for TiO_2 to eliminate methylene blue. The calcium alginate/ TiO_2 beads were prepared through the immobilization method. A fixed proportion of the reactor was filled with the catalyst for efficient mass transfer. Moreover, alginate beads act as a semipermeable membrane that permits the passage of low molecular weight water-soluble molecules. Albarelli et al. (2009) also explained that methylene blue degradation follows initial adsorption followed by photocatalytic degradation. The alginate beads also exhibited very good reusability property up to 20 cycles because of enhanced superficial area. In another study, laccases were used along with alginate due to their high oxidizing ability. The spore laccase/ TiO_2 nanoparticles entrapped alginate beads were reported to eliminate the 25 mg/L of indigo carmine. The studies confirmed 69% indigo carmine removal under the illumination of a light source within 60 min (Khakshoor et al. 2021).

Wei et al. (2016) described the preparation of polyacrylamide/calcium Alginate/ TiO_2 composite hydrogel film of specific thickness 0.3 mm under UV irradiation and Ca-crosslinking. The prepared film demonstrated superior mechanical strength and toughness. The photocatalytic studies revealed that 81% methyl orange was removed within a contact time of 120 min under ultraviolet illumination with reusability of 5 cycles. Gjjipalaj & Alessandri, (2017) prepared alginate-based TiO_2 beads through ionotropic gelation for the removal of both anionic (Methyl orange) and cationic dye (Methylene blue). The beads morphology of alginate-based TiO_2 adsorbed a higher amount of dyes than in powder form. However, beads took more time than powder for methylene blue removal due to the porous catalysts reduced surface area. The beads also exhibited reusability up to five consecutive cycles under ultraviolet light irradiation. So, the key advantages of preparing beads are their reusability and ease of recoverability.

Kanakaraju et al. (2017), described the dual usage of TiO_2/ZnO -calcium alginate beads to remove copper through

adsorption and photocatalysis, respectively. The immobilized beads were prepared with three different mass ratios of TiO_2 and Zinc (1:1, 1:2, 2:1) to redeem pollutants under direct ultraviolet light. The photocatalytic studies revealed that these beads (1:1) could remove 7.14 to 62.0 mg/L of copper (initial concentration of 10 to 80 mg/L). The better efficiency of this catalyst may be due to the enhanced adsorption of the metal cations on the surface of beads. Later, magnetic TiO_2 /Alginate/Fe nanoparticles triads were developed via facile non-thermal method. The unique property of alginate in this prepared catalyst is their ability to act as an obstacle between the direct contact of magnetic core and TiO_2 . This magnetic catalyst also displayed 97.6% of methylene blue removal within 120 min under the illumination of ultraviolet light (254 nm). The efficiency of the prepared catalyst was reduced to 88.5% after three consecutive cycles (Kanakaraju et al. 2018).

Reveendran and Ong (2018) focused on removing dyes through the development of sodium alginate- TiO_2 films. The catalyst demonstrated more than 50% removal of Congo red. Similarly, the sodium alginate- TiO_2 -bentonite nanocomposite was prepared through in-situ method for the reclamation of methylene blue from wastewater. The degradation studies showed 90.5% removal of methylene blue at the optimized condition (pH 8, time 30 min, and catalyst dosage of 30 mg/L) (Subekti and Helmiyati 2018). In the highly stable and porous polysulfone-alginate/ TiO_2 photocatalyst, the polysulfone matrix resembled as a sponge-macro-void structure; alginate anchored the sites of TiO_2 nanoparticles and restricted the surface porosity. TiO_2 enhanced the thermal and mechanical properties of the beads. The polysulfone-alginate/ TiO_2 displayed complete removal of methylene blue, triclosan, diclofenac within 50, 40, and 120 min. Figure 4A shows the photocatalytic redemption of diclofenac and triclosan by utilizing the polysulfone-alginate/ TiO_2 . The appearance and disappearance of the colored by-products of diclofenac at different time intervals are shown in Fig. 4B. The stability of the beads is also observed in Fig. 4C. The enhanced removal rate is attributed to the catalysts higher surface area and greater adsorption properties. The catalyst ensured reusability for 25 consecutive cycles with a 99% removal rate (Mehmood et al. 2020). Floating TiO_2 /polyvinyl alcohol/alginate composite exhibited a 28.5% and 29.3% ammonia degradation under visible and ultraviolet light, respectively. The floatable catalyst displayed reusability up to 5 cycles (Zendehzaban 2019).

Dalponte Dallabona et al. (2021) prepared floating catalyst with Brazilian bentonite/ TiO_2 /alginate beads by adopting the gelation method. Prepared beads exhibited spherical shape, and the addition of bentonite does not influence the size. However, the addition of gas-forming agents (calcium carbonate) increased the average diameter of floating catalyst from 3 to 4.9 mm. The significant enhancement in size was

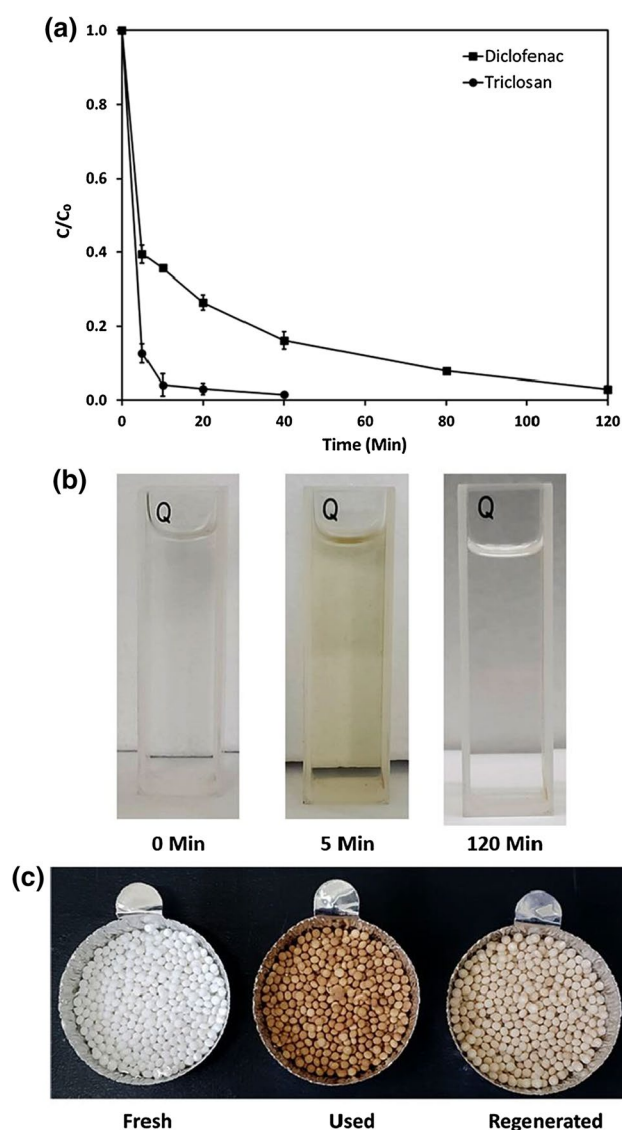


Fig. 4 **A** Photocatalytic degradation of diclofenac and triclosan using polysulfone-alginate/TiO₂, **B** Photographs of diclofenac sample yielded at different time intervals showing the formation of different colored by-products, **C** The photograph images of polysulfone-alginate/TiO₂ beads before and after use. The stability of the photocatalyst is also understood from the figure in detail. Reprinted from Mehmood et al. (2020) with permission from Elsevier

also attributed to the release of carbon dioxide with carbonate decomposition in an acidic-acetic base. Moreover, the surface area of the beads was 4.447 m²/g, greater than that of sodium bentonite (3.562 m²/g). Photocatalytic studies of bentonite/TiO₂/alginate beads demonstrated a 99% dye removal with a total organic carbon removal of 34% and 53% for tartrazine and methylene blue, respectively. Floating catalyst also exhibited a reusability up to 7 cycles and yielded a degradation of > 80%.

Mallakpour et al. (2019) reported the sono-assisted synthesis of the alginate/carbon nanotube/carbon dot/

fluoroapatite/TiO₂ beads for the elimination of methylene blue from wastewater. Figure 5 shows the schematic representation of different stages during the preparation of alginate/carbon nanotube/carbon dot/fluoroapatite/TiO₂ beads. Studies reported the higher efficiency of the prepared photocatalyst under the illumination of ultraviolet light. The presence of the carbon nanotubes, carbon dot, and fluoroapatite not enhanced the removal efficiency but also minimized the recombination of photo-induced electron–hole pairs. The hydroxyl radical’s superoxide radicals and holes played a phenomenal role in degradation of dyes. Recently, Chkirida et al. (2021) prepared alginate/bentonite impregnated TiO₂ beads to degrade methylene blue present in the wastewater by utilizing a green and simple impregnation method. The obtained photocatalyst showed 98% methylene blue and 93% chemical oxygen demand reduction within one hour of light irradiation. Table 4 summarizes different alginate-supported TiO₂ photocatalysts for removing organic pollutants.

Alginate-based materials are widely used in developing multifunctional systems, particularly for photocatalytic applications. The uniqueness of alginate is ascribed to biocompatibility, stability, changeable porosity, and low cost. Even after continuous research, the following points must be addressed as earliest to attain a sustainable solution.

(i) Sodium alginates mechanical strength and stability are determined to be relatively poor. As a result, additional attention must be paid to the development of alginate-based polymers with increased surface area and active sites.

(ii) Developing a low-cost, effective synthesis process is essential for the catalyst’s enhanced structure.

(iii) Most studies reported the reusability of the alginate-based materials up to 5 cycles only. Thus, developing a highly stable alginate-based photocatalyst is mandatory for large-scale applications.

Cellulose

Cellulose is the most abundant natural polymer obtained from cotton, rice husks, banana, acetobacter, rhizobium, aerobacter, pseudomonas, and sarcina. Cellulose is a tough and insoluble fiber due to strong inter and intramolecular hydrogen bonds that always benefit the maintenance of plants cell walls (Mohamed et al. 2017). The unique properties of cellulose are high flexibility, biodegradability, and protecting capability (Li et al. 2014). Different materials including cellulose acetate, cellulose nanofibers, and nanocrystalline cellulose was produced from cellulose sources (Mohamed et al. 2017).

Cellulose acetate is an ester of cellulose derived through the chemical reaction between cellulose with the anhydride of acetic acid in the presence of sulfuric acid. Cellulose acetate can be drawn into beads, sheets, films, membranes, and nanocomposites. The cellulose acetate is preferred due

to high electrospinning ability, ease of dilution, porosity, better surface area, and good hydrolytic stability (Jatoi et al. 2019). The potent usage of cellulose acetate is extended to textile industries, water treatment, adsorption, pharmaceuticals, and membranes.

Mohamed et al. (2015) emphasized the development of nitrogen-doped TiO₂ nanorods in regenerated cellulose thin film through the phase inversion method. The recycled newspaper was used as a sustainable cellulose source throughout this study. The prepared catalyst exhibited a remarkable property between the cellulose and TiO₂ nanorods anchored on the thin film, which also resolved the problems related to aggregation of TiO₂ photocatalyst. The optical studies confirmed the introduction of localized N 2p states just above the valence band due to nitrogen doping resulting in the enhancement of visible light absorption. Moreover, the bandgap of the thin film was reduced from 3.09 to 2.95 eV for the prepared thin film. The prepared catalyst demonstrated 96% and 78.8% methylene blue removal under ultraviolet and visible light illumination.

Differently, Marinho et al. (2017) prepared the cellulose acetate monoliths coated with TiO₂ utilizing the dip-coating method. Here, cellulose acetate monoliths were employed due to their higher irradiated surface area inside the reactor, increasing light utilization. The prepared structure exhibited a better-illuminated surface area of 212 m²/m³. Experimental investigations reported only 10% chromium (VI) removal within 210 min under the absence of citric acid. Even though studies reported the chromium (VI) reduction by single-electron steps, gradual reduction and re-oxidation of chromium species occurred due to the conduction band electrons and valence band holes or hydroxyl radicals. The cellulose acetate monoliths coated with TiO₂ exhibited reusability up to 10 consecutive cycles.

The elimination of the hydrophobic compounds using cellulose-based materials is restricted due to cellulose's hydrophilic characteristics. This particular limitation is improved by a team of researchers led by Ríos-Gómez et al. (2018) through chemical modification. This resulted in the enhancement of the interaction between catalyst and pollutant. Ríos-Gómez et al. (2018) also explored cellulose as a base substrate for the development of TiO₂ based nanocomposite film for removal of methyl orange. The cellulose paper was coated with polyamide/TiO₂ photocatalyst through the simple dipping method. Under the illumination of ultraviolet light, the photocatalyst degraded methyl orange completely within 20 min. The presence of cellulose improved the mechanical stability and acted as an efficient sorptive phase against the pollutants. The polymeric nature of the photocatalyst attributed to the stabilization of TiO₂ nanoparticles and exhibited higher ability towards the retention of the pollutant, which favors photocatalytic degradation of methyl orange. In the same year, Li et al. (2018) focused

on overcoming the poor mechanical stability of cellulose nanofibers via chemical crosslinking. Cellulose nanofiber/TiO₂ aerogel photocatalyst was chemically cross-linked via hydrothermal method. The TiO₂ particles have adhered to the surface of cellulose nanofibers as scaffold through a hydrogen bond. Photocatalytic studies proved the potential viability of catalysts in the redemption of methylene blue under irradiation of ultraviolet light. The catalyst also extended the reusability to five cycles and exhibited 72% at the end.

Shi et al. (2019) also used TiO₂ doped chitosan microspheres supported cellulose acetate fibers to remove methyl orange. The cellulose acetate material was used as base material to increase the water flux of the membrane. Moreover, the chitosan hemisphere gradually dispersed TiO₂ nanoparticles and helped to enhance the catalyst surface. At optimum conditions of pH 4, and initial concentration = 40 mg/L, about 98% methyl orange removal was observed. Even at the end of the fifth cycle, the membrane exhibited 98% degradation efficiency due to the presence of chitosan in the membrane, which made the intermolecular hydrogen bonds stronger among chitosan, cellulose acetate and TiO₂. In the same year, ZabihiSahebi et al. (2019) synthesized cellulose acetate/chitosan/single walled carbon nanotube/Fe₃O₄/TiO₂ composite nanofibers using the electrospinning method. The presence of single walled carbon nanotubes increases the removal efficiency due to their ability to disperse at a very low concentration. Catalyst also displayed a phenomenal activity towards the reduction of chromium (VI), arsenic (V), methylene blue, and congo red from wastewater.

For instance, Chen et al. (2020a, b) emphasized the eco-friendly cellulose/graphene oxide/TiO₂ hydrogel photocatalyst synthesized through one-step green and simple method for removing methylene blue. The catalyst exhibited 93% methylene blue removal within 120 min with reusability up to ten consecutive cycles. The enhanced degradation efficiency was attributed to the stronger interaction between TiO₂ and graphene oxide, facilitating interfacial charge transfer and minimizing the electron-hole recombination. Moreover, the higher surface area of the cellulose hydrogel provided a large number of active sites and enhanced the surface adsorption of methylene blue. These hydrogels composed of graphene oxide and TiO₂ maintained the hydrogel structure and did not cause secondary pollution.

Yang et al. (2020) focused on the preparation of three-dimensional bacterial cellulose/polydopamine/TiO₂ membrane adopting a facile strategy. Figure 6a–d describes the surface morphology of the bacterial cellulose/polydopamine/TiO₂ membrane. Studies indicated that the uniform dispersion of TiO₂ nanoparticles on the bacterial cellulose/polydopamine matrix. This also played a critical role in enhancing the surface area of 170 m²/g. The prepared

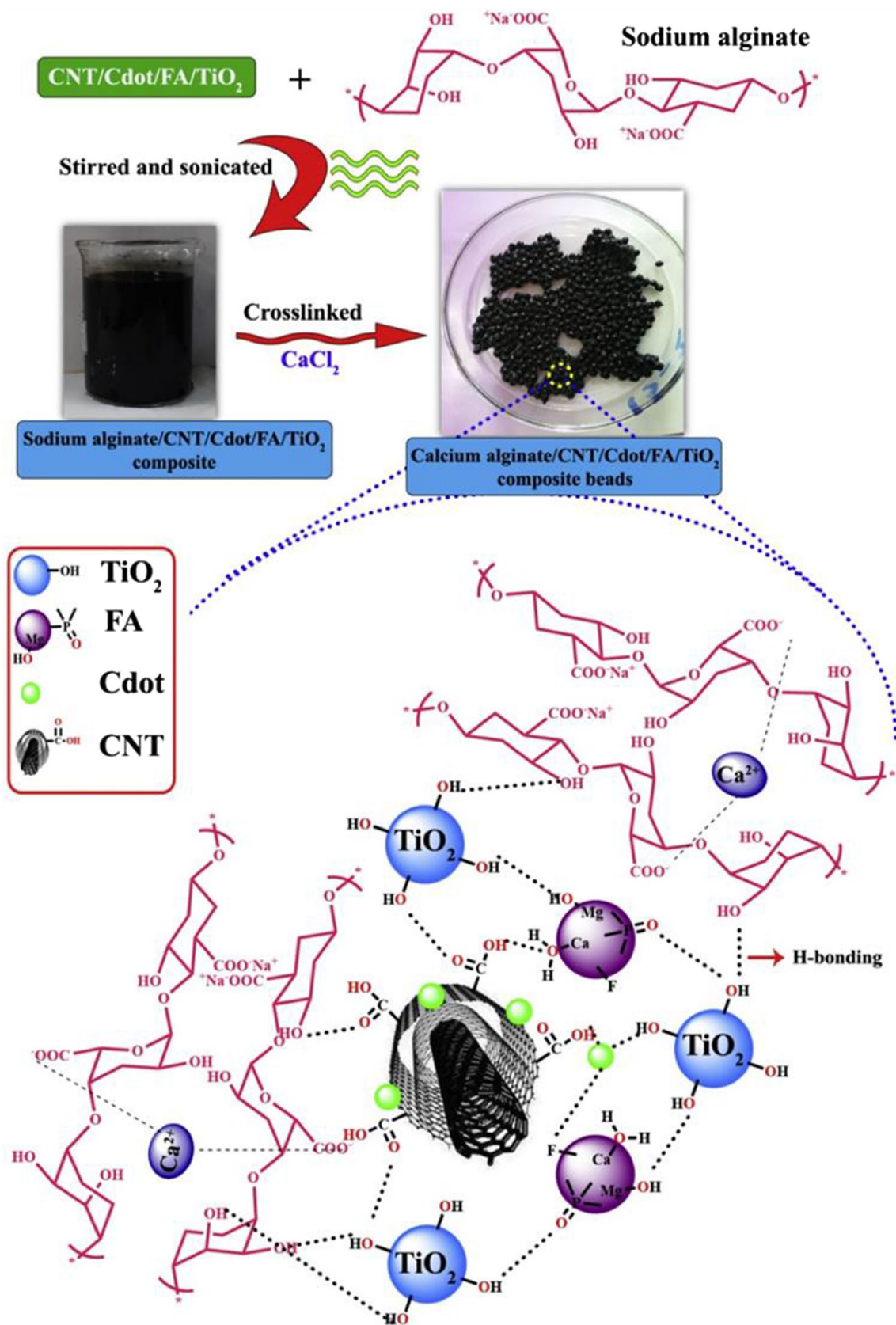


Fig. 5 Preparation mechanism of floatable with alginate/carbon nanotube/carbon dot/fluoroapatite/TiO₂ beads. The key steps involved in the preparation are (i) Mixing of carbon nanotube/carbon dot/fluoroapatite/TiO₂ with sodium alginate solution via sonication. (ii) Drop the mixture into a calcium chloride bath using a syringe to yield beads. Finally wash the prepared beads to eliminate the excess calcium chloride on the bead surface. Reprinted from Mallakpour et al. (2019) with permission from Elsevier. Where FA is fluoroapatite, Cdot is carbon dot, and CNT is carbon nanotube

composite membrane could remove 100% of rhodamine B, 99.5% of methylene blue, and 95.1% of methyl orange within 60, 20, and 30 min under the illumination of a 500 Watts Hg lamp. The prepared composites demonstrated reusability up to five consecutive cycles with a reduction in 5% removal efficiency as observed in Fig. 6e. Thus, incorporating cellulose has improved the adsorption stability, reusability, and photocatalytic performance of the photocatalyst. Moreover, the polydopamine acted as a secondary reaction route by forming a non-covalent bond with TiO₂ particles. The polydopamine also boosted the catalytic efficiency through promoting photo-induced charge-carrier species at the interface between polydopamine and TiO₂.

Recently, Lin and Huang (2021) synthesized the H₃PW₁₂O₄₀/TiO₂ with cellulose as a scaffold, which acted as a template with a porous structure. The H₃PW₁₂O₄₀ has served as an effective electron acceptor and improved the separation of electron–hole pairs in eliminating methylene blue and rhodamine B under sunlight. Experimental studies proved that the photocatalyst developed through impregnation method was better than the layer-by-layer method due to the closer contact between the TiO₂ and H₃PW₁₂O₄₀ in cellulose matrix. Additionally, the catalyst prepared through the impregnation method can act as an effective photocatalyst towards the elimination of pollutants under direct sunlight. In the same year, Wafiroh et al. (2021) reported the preparation of cellulose acetate hollow fiber membrane from banana stems coated with TiO₂. The highly porous structure of the prepared hollow fiber membrane showed the presence of rigid pores with enhanced mechanical properties. The photocatalytic studies showed a removal efficiency of 94.96% for congo red and 94% for textile wastewater. The studies on reusability are necessary to showcase the stability of the material.

Table 5 summarizes different cellulose-supported TiO₂ photocatalysts for the removal of organic pollutants. The TiO₂/cellulose-based structures always lead the path for the preparation of novel photocatalytic materials. The following benefits are observed with the usage of cellulose as a support material (Hamad et al. 2020).

- Control the hydrolysis, polycondensation, and growth of cellulose/metal oxides
- Provides strong mechanical support by acting as an attractive material for the suspension of the photocatalyst.
- Tune the optical properties of photocatalyst, shifts the absorption from ultraviolet to visible light region.
- Improves electron distribution and electron transfer to the catalyst surface and restricts the recombination of electron and hole pairs.
- Improves the thermal stability of the photocatalyst and enhance adsorption in the absence of light
- The cellulose-supported membrane sheets also make the recovery and reusability easier.

The stability and mechanical properties of cellulose-based TiO₂ materials can be increased by altering their proportions. The development of different morphologies of cellulose-based materials is important to overcome the issues related to separation and recovery. The detailed study on the degradation mechanism and the photocatalytic process for various pollutants are beneficial in designing suitable materials. In addition, the detailed analysis on pollutant removal and recycling of materials is necessary for large-scale wastewater treatment plants. The development of cellulose-based composites with high performance and selectivity is a great barrier to practical applications.

Cyclodextrin

Antoine Villiers firstly reported cyclodextrin in the nineteenth century (Liu et al. 2019). Cyclodextrin was initially used in pharmaceuticals, food industries, and chromatography and later conquered different areas in the field of drug delivery, and water treatment (Adeoye & Cabral-marques, 2017; Liu et al. 2019; Preethi et al. 2020; Tian & Liu, 2020). Cyclodextrin is a cyclic low molecular oligo polymer yielded from the starch via enzymatic action (Crini et al. 2018; Sherje et al. 2017). Cyclodextrin are composed of six to eight units of α -D-glucose represented as denoted α -, β -, and γ -cyclodextrin (Rajalakshmi et al. 2019; Sherje et al. 2017; Tian & Liu, 2020). Literature reported that cyclodextrin forms an inclusion complex with different molecules of appropriate polarity and dimension with hydrophobic interior and hydrophilic external surface (Rajalakshmi et al. 2019; Tian and Liu, 2020). The cyclodextrin are hollow, truncated cone-shaped molecules with a large number of glucose units connected with oxygen covalently through hydrogen bonds (Crini et al. 2018). Cyclodextrin are frequently used because of the low cost, availability, complex formation towards large complexes (Crini et al. 2018).

The cyclodextrin-supported polymers, semiconductor-based materials, metals, carbon nanomaterials have been reported as sustainable and efficient materials for wastewater

Table 4 Sodium alginate-supported TiO₂ photocatalysts

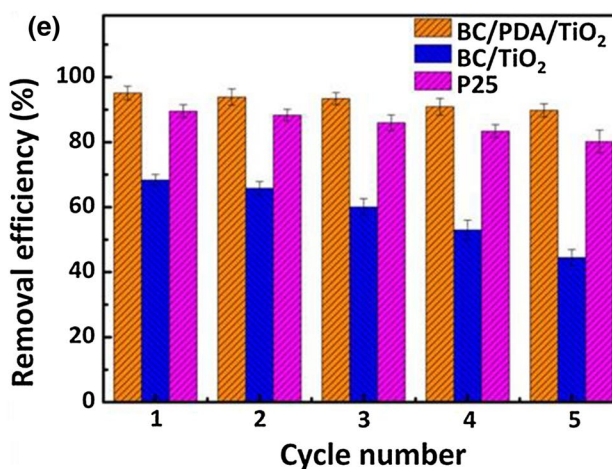
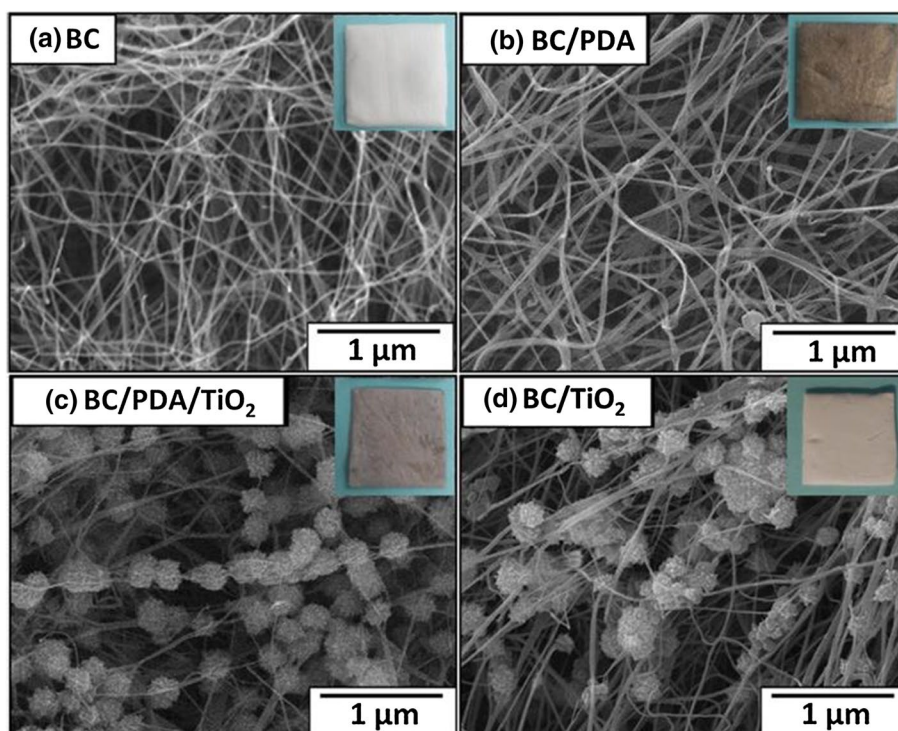
Catalyst	Pollutant	Light source	Initial concentration (mg/L)	Time required (minute)	Efficiency (%)	References
Reduced graphene oxide-TiO ₂ /sodium alginate 3-D aerogel	Ibuprofen, Sulfamethoxazole	High-pressure mercury lamp	10	90	78 99	Nawaz et al. (2020)
TiO ₂ /calcium alginate photobiocomposites	Basic blue 442	Direct sunlight	30	240	96	Nouri et al. (2020)
Novel TiO ₂ /calcium alginate floating catalyst	Tartrazine dye	Ultraviolet (125 Watt)	50	180	88.97	Dalponte et al. (2019)
Alginate/carbon nanotube/carbon dot/fluoroapatite/TiO ₂ beads	Methylene blue	4 lamps of ultraviolet-C lamp (8 Watt)	10	315	–	Mallakpour et al. (2019)
TiO ₂ /calcium alginate beads	Basic red 18, reactive red 180	Ultraviolet-A (8 Watt)	25	180	87.1 45.6	Isik et al. (2019)
Graphene oxide-TiO ₂ /sodium alginate reduced graphene oxide-TiO ₂ /sodium alginate aerogels	Microcystin-LR	Ultraviolet	–	120	–	Nawaz et al. (2018)
Graphene oxide and TiO ₂ nanoparticles incorporated alginate/carboxymethyl cellulose Nanocomposites	Congo red	Sunlight	30	240	98	Thomas et al. (2016)
Floatable TiO ₂ /polyvinyl alcohol-sodium alginate composites	Ammonia waste water	High-pressure mercury lamp (250 Watt)	200	120	63	Zendehzaban (2019)
Immobilized TiO ₂ in calcium alginate beads	Ibuprofen, atenolol, carbamazepine	Ultraviolet-A	–	–	99	Sarkar et al. (2015)
Calcium alginate/TiO ₂ beads	Methylene blue	Ultraviolet (9 Watt)	20	1440	> 95	Lam et al. (2017)
TiO ₂ /alginate/Fe nanoparticles	Cr (III) Cu (II) Pb (II)	Ultraviolet-C (120 Watt)	44	72	98.6 98.4 99.5	Kanakarajuet al. (2019)
TiO ₂ -polyvinyl alcohol-alginate beads	Pb (II)	Natural sunlight	50	150	98	Idris and Majidnia (2014)
TiO ₂ /calcium alginate composite film	Methyl orange	Ultraviolet	–	120	82.25	Zhao et al. (2013)
Maghemite and titania nanoparticles in polyvinyl alcohol-alginate encapsulated beads	Pb (II)	Sunlight	50	100	100	Majidnia and Idris (2015)

remediation through photocatalysis and adsorption (Kumari 2020). The cyclodextrin showed an extremely good adsorption property for organic contaminants, bringing them close to the photocatalyst surface and enhancing the removal efficiency. Further, the catalysts offer very high catalytic efficiency, durability, and stability and are used as an alternative towards commercial TiO₂. Finally, cyclodextrin-supported materials have a promising future in the field of wastewater

treatment because of low energy requirements and trap and zap treatment methods (García-díaz et al. 2020).

Zhang et al. (2020) emphasized the utilization of Ag/ β -cyclodextrin co-doped TiO₂ floating photocatalytic membrane synthesized via modified electrospinning process coupled with the low-temperature in-situ hydrothermal method. The Brunauer–Emmett–Teller results revealed that the surface area of the catalyst increased from 0.0192

Fig. 6 Surface morphology **a** Interwoven three dimensional nanofibrous structure of the pristine bacterial cellulose membrane of 40 nm in diameter, **b** In polydopamine/bacterial cellulose, the diameter of nanofibers increased to 50 nm, **c** The increment of size of nanofibers is enhanced to 200 nm for the bacterial cellulose/polydopamine/TiO₂, **d** The spherical TiO₂ particles are evidently seen in the bacterial cellulose/TiO₂ matrix. **e** Reusability of the photocatalysts. A decline of 5.5%, and 23.8% in the removal efficiency is seen for bacterial cellulose/polydopamine/TiO₂ and bacterial cellulose/TiO₂, respectively. Reprinted from Yang et al. (2020) with permission from Elsevier. Where BC is bacterial cellulose, and PDA is polydopamine



to 0.0652 cm³/g. The existence of mesopores is the key factor in enhancing the adsorption behavior of the photocatalyst. The incorporation of Ag, cyclodextrin with TiO₂ altered the bandgap to 1.9 eV and was active under visible light. The Ag/β-cyclodextrin co-doped TiO₂ degraded *N,N*-dimethylformamide under visible light (150 Xenon lamp). The photocatalytic degradation mechanism of the prepared catalyst is illustrated in Fig. 7. The floating photocatalyst combine with the visible light and oxygen directly. Because of the surface plasmon resonance effect, the Ag nanoparticles near their plasmon frequency exhibit a trend of being excited by visible light and generate electron field and produce the electron–hole pairs. The transfer of electrons to the conduction band of TiO₂ and were trapped by the oxygen molecules in the mesopores to form superoxide radicals. The

hole scavenging properties of the C, photo-induced holes transfer from valence band of TiO₂ to cyclodextrin and minimize the recombination of electron–hole pairs. The floating photocatalytic membrane has potential significance in wastewater treatment because of extended reusability, durability, mechanical strength, hydraulic permeability.

The cyclodextrin-directed synthesis of Au/TiO₂ materials was prepared through the colloidal self-assembly approach under visible light illumination. Moreover, the catalyst removed the phenoxyacetic acid from aqueous streams. Reusability studies demonstrated a decline in catalytic activity from 85 to 52% during the third run. The decline in the catalytic activity is ascribed to the competitive adsorption and agglomeration of the phenoxyacetic acid on the catalyst surface through the hydrogen bond formation between

Table 5 Cellulose acetate supported TiO₂ photocatalyst

Catalyst	Pollutant/Initial concentration	Light source	Contact time (min-ute)	Efficiency (%)	Reusability	References
Bimetallic Au/Ag decorated TiO ₂ nanocomposite membrane	Tetracycline (5 mg/L)	Xenon lamp	120	80 (static) 90(continuous)	–	Li et al. (2019)
Cellulose acetate monoliths structure coated with TiO ₂	Chromium (VI) (0.01–0.06 mM)	Xenon arc lamp	–	–	10	Marinho et al. (2017)
Polyvinylpyrrolidone/cellulose acetate/TiO ₂ /nanofibrous membrane	Methylene blue (10 ⁻⁵ M)	Ultraviolet	–	–	–	Orlando et al. (2020)
TiO ₂ -doped chitosan microspheres supported on cellulose acetate	Methyl orange (40 mg/L)	Mercury lamp (25 Watt)	0 to 85	> 98	5	Shi et al. (2019)
Polyacrylonitrile/cellulose acetate/MIL-125/TiO ₂ composite	Dozorubicin (50 mg/L), chromium (VI) (50 mg/L)	4 ultraviolet lamps (30 Watt)	60	> 90	–	Bahmani et al. (2020)
Carbon-TiO ₂ doped cellulose nanocomposite film	Reactive red-195 (20 mg/L)	High pressure mercury lamp (125 Watt)	420	99.5	4	Pham et al. (2020)
H ₂ O ₂ assisted TiO ₂ and micro cellulose composites	Methylene blue (200 mg/L) acid violet (200 mg/L) methyl violet (200 mg/L)	Solar light	150 360 420	99 96 95	4 for methylene blue	Rajagopal et al. (2020)
Microfibrillated cellulose/TiO ₂ films	Methylene blue (5 mg/L)	Ultraviolet lamp (10 Watt)	90	90	–	Ng and Leo (2019)
TiO ₂ /Cellulosic fiber	Paracetamol (2.65 × 10 ⁻⁴ M)	Low-pressure mercury lamp (11 Watt)	150	> 90		Jallouli et al. (2017)
Microfibrillated cellulose-polyamide-amineepichlorohydrin-TiO ₂ nanoparticles	Methyl orange	Solar light	150	83 95	5 3	Garusinghe et al. (2018)
Microcrystalline cellulose/carrageenan/TiO ₂ film	Methylene blue	Ultraviolet lamp	600	83	–	Jo et al. (2017)
Regenerated cellulose/N-doped TiO ₂ nanocomposite membrane	Phenol (40 mg/L)	Ultraviolet (30 Watt)	360	96.8	–	Mohamed et al. (2016)

surface hydroxyls and carboxylic acid groups of phenoxyacetic acid reduce the photonic efficiency (Lannoy et al. 2017). In the same year, Zhu et al. (2017) reported the deposition of cyclodextrin/gold nanoclusters on TiO₂ nanoparticles. The prepared material could remove 98% of methyl orange in 10 min. The high removal efficiency is attributed to the unique cavity of the prepared catalyst improved the

adsorption through host-guest interaction. The Au in the core helped in accepting the electrons and minimized the recombination rate of electron/hole pairs.

Recently, Zhang et al. (2021) focused on preparing TiO₂/graphene oxide on polyacrylonitrile/ β -cyclodextrin nano-fibrous membrane by ultrasonic-assisted electro-spray method. Experimental investigations found that the

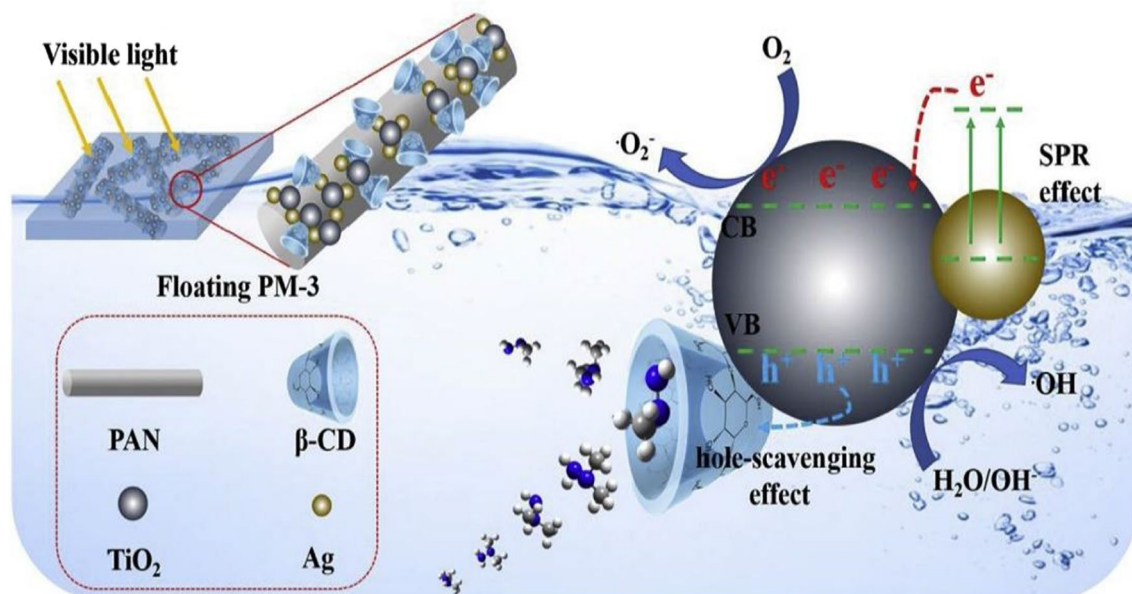


Fig. 7 Degradation mechanism of Ag/ β -cyclodextrin co-doped TiO_2 floating photocatalytic membrane. The photocatalytic membrane float in the surface of water and combine with the incident visible light and oxygen directly. Because of the surface plasmon resonance, Ag nanoparticles are excited by visible light and produce an electric field and results in generation of hot electron-hole pairs. These electrons are transferred to conduction band of TiO_2 and trapped by absorbed O_2 in the mesopores to form superoxide radicals. The hole scavenging

effect of cyclodextrin resulted in the transfer of holes from valence band of TiO_2 to cyclodextrin. This also leads to minimized recombination of electron-hole pairs. Reprinted from Y. Zhang et al. (2020) with permission from Elsevier. Where β -CD is β -cyclodextrin, Ag is silver, PAN is polyacrylonitrile, SPR surface plasmon resonance, PM is photocatalytic membrane, e is electron, h is hole, CB is conduction band, and VB is valence band

prepared material exhibited 93% and 90% removal efficiency for methylene blue and methyl orange, respectively. Moreover, this membrane displayed good antibacterial properties and extended reusability up to 3 cycles. The inclusion of polyacrylonitrile and cyclodextrin into the structure of the desired photocatalyst (i) impart low density and hydrophobicity to the catalyst, which could enhance the light penetration, (ii) minimize the problems due to the secondary pollution caused by the leaching of the TiO_2 and graphene-oxide from the surface of the photocatalyst, (c) enhance porosity and specific surface area of the nanofiber.

Similarly, Wang et al. (2021) focused on removing bisphenol-A from aquatic streams by utilizing the TiO_2 @ aspartic acid- β -cyclodextrin@ reduced graphene oxide developed through photochemical method as highlighted in Fig. 8a. The surface morphology showed uniform distribution of TiO_2 nanoparticles on the catalyst surface due to the inclusion of aspartic acid (enhancing water solubility). The prepared catalyst displayed high degradation efficiency towards eliminating 5 mg/L of bisphenol-A under the illumination of the ultraviolet light (250 Watts metal halide lamp). Moreover, O_2^- and holes were responsible for the elimination of bisphenol-A (see Fig. 8b). The possibility of recombining photogenerated electron-hole pairs on catalyst surfaces may lead to low quantum yield. The higher

charge separation and electron transfer are necessary to benefit greater efficiency. The catalyst exhibited higher stability and reusability up to 5 consecutive cycles. The presence of the cyclodextrin enhanced the mass transfer process at the interface and enhanced the photocatalytic activity. At the same time, graphene triggered the electron transfer during the degradation process.

Table 6 summarizes different cyclodextrin-supported TiO_2 photocatalysts for the removal of organic pollutants. Cyclodextrin is a versatile material for sustainable treatment technologies and can remove various pollutants. Even though cyclodextrin-based materials exhibit higher efficiency, further studies are required to overcome the hindrances or the application towards large-scale treatment plants. The eco-friendly routes need to be identified to synthesize cyclodextrin/ TiO_2 based materials.

Other biopolymer-supported TiO_2 catalysts

Starch

Starch is a bio-polymer abundantly available from wheat, paddy, potatoes, corn (Sarmah and Karak 2020). Starch is a three-dimensional structure with better crystallinity (15 to 45%) and possesses D-glucose units with macromolecules

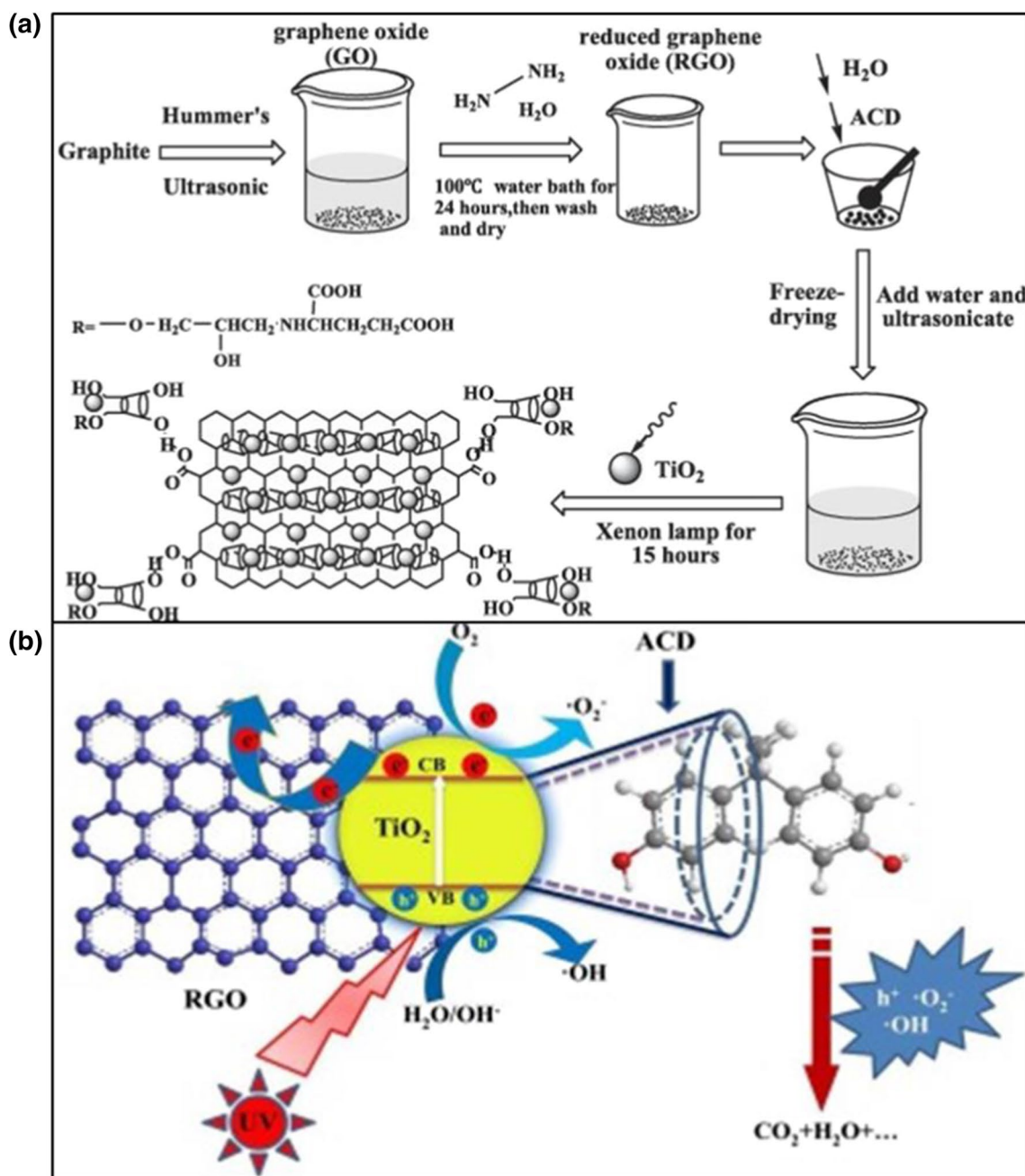


Fig. 8 **a** Preparation of TiO_2 @aspartic acid- β -cyclodextrin@ reduced graphene oxide. The aspartic acid- β -cyclodextrin@ reduced graphene oxide was dispersed in water followed by the ultrasonic treatment. To this solution TiO_2 was added with continuous stirring and exposed to simulated sunlight for 15 h and dried in vacuum at 65°C . **b** Photocatalytic degradation mechanism. The introduction of reduced graphene oxide to the photocatalytic system, caused the electron transfer from semiconductor to reduced graphene oxide. This suppresses the

recombination of electron-hole pairs. The inclusion of aspartic acid- β -cyclodextrin can form an inclusion complex with bisphenol-A and enhance the mass transfer of bisphenol-A at the photocatalyst surface and enhance the catalytic activity. Reproduced with permission from Wang et al. (2021) with permission from Elsevier. Where GO is graphene oxide, RGO is reduced graphene oxide, e is electron, h is hole, CB is conduction band, VB is valence band, UV is ultraviolet, and ACD is aspartic acid- β -cyclodextrin

similar to amylopectin, amylose, and glucon. The nanocrystal starch and microcrystalline starch are yielded through the hydrolysis (Bahrami et al. 2019). The popularity of the starch derivatives increased due to the higher surface area,

abundance, low cost, non-toxicity, and renewability. For example, Guo et al. (2019) also focused on preparation of synthesizing TiO_2 cross-linked carboxymethyl starch composites by utilizing corn as a natural source through sol-gel

Table 6 Cyclodextrin-based TiO₂ photocatalysts

Catalyst	Pollutant/Initial concentration	Light source	Contact Time (min)	Efficiency (%)	Reusability	References
Ti ³⁺ -doped TiO ₂ composited with NiO and β -cyclodextrin	Bisphenol-A (20 mg/L)	Xenon lamp (500 Watt)	90	95	5 (95%)	Wang et al. (2020)
TiO ₂ microspheres with cross-linked cyclodextrin	Tetracycline (50 mg/L)		20	100		
	Rhodamine-B (20 mg/L)		10	100		
	Bisphenol-A (20 mg/L)	6 Ultraviolet-A lamp (4 Watt)	180	80	4 (71.7%)	García-díaz et al. (2020)
Novel TiO ₂ @aspartic acid- β -cyclodextrin@ reduced graphene oxide	Bisphenol-A (5 mg/L)	Metal halide lamp (250 Watt)	60	85.6	5 (76.4%)	Wang et al. (2021)
Cyclodextrin grafted MoS ₂ /g-C ₃ N ₄	Glyphosate (9 g/L)	Sunlight	180	–	5	Wu et al. (2020)
TiO ₂ - β -cyclodextrin	Chromium (VI) (10 mg/L)	Sunlight	180			
	Methyl orange (5 mg/L)	Visible light (500 Watt)	120	89.4	–	Sakthivel and Velusamy (2017)
	Methyl orange (10 mg/L)	Natural sunlight	300	90.92	3 (above 80% for both)	Zhang et al. (2021)
Polyacrylonitrile/ β -cyclodextrin composite nanofibrous membranes immobilized with TiO ₂ and graphene oxide						
Carboxymethyl- β -cyclodextrin-Fe ₃ O ₄ @TiO ₂	Methylene blue (10 mg/L)		300	93.52		
	Polychlorinate biphenyls	Ultraviolet lamp (8Watt)	16	83	5	Khammar et al. (2020)
Ag/ β -cyclodextrin co-doped TiO ₂ floating photocatalytic Membrane	Unsymmetrical dimethylhydrazine (20 mg/L)	Xenon lamp (150 Watt)	80	96.8	5 (Not much change)	Zhang et al. (2020)
Wood-flour/cyclodextrin/TiO ₂ hybrid composite	Methyl orange (0.1 mmol/L)	High pressure mercury lamp (250 Watt)	–	100	5 (86%)	Sun et al. (2016)
Cyclodextrin-functionalized Fe ₃ O ₄ @TiO ₂ magnetic nanoparticles	Bisphenol-A (20 mg/L), dibutyl phthalate (20 mg/L)	Mercury vapor lamp (400 Watt)	175	98	10 (90%)	Chalasan i and Vasudevan (2013)
Ag and β -cyclodextrin co-doped TiO ₂ nanoparticles loaded onto activated carbon	Naphthalene	Visible light	150	98.4%	–	Chen et al. (2017a, b)
carboxymethyl β -cyclodextrin-P25	Sulphanilamide (20 mg/L)	Mercury lamp (10 Watt)	60	97.1	5 (> 90%)	Zhou et al. (2020)
	Phenol (20 mg/L)			71.9	–	
β -cyclodextrin-functionalized carbon nanotubes/TiO ₂	Bisphenol-A (20 mg/L)			91.6	–	
	Rhodamine B (20 mg/L)	Xenon lamp (150 Watt)	180	86.7	–	Zhang et al. (2019)
	Methylene blue (20 mg/L)		180	97	5 (93%)	
	Congo red (20 mg/L)		180	88.1		

Table 6 (continued)

Catalyst	Pollutant/Initial concentration	Light source	Contact Time (min)	Efficiency (%)	Reusability	References
β -Cyclodextrin grafted titanium dioxide	2,4-dichlorophenoxyacetic acid (20 mg/L)	Light emitting diode (20 Watt)	300 (batch)	95	5 (84%)	Safa et al. (2019)
Ag-titania nanotubes/magnetic chitosan/ β -cyclodextrin	Chrome black T (20 mg/L)	Xenon lamp	480 (continuous)	96	-	Ding et al. (2017)
TiO ₂ / β -cyclodextrin	Rhodamine B	Sunlight	120	-	-	Rajalakshmi and Pitchaimuthu (2017)
Carboxymethyl- β -cyclodextrin functionalized La@TiO ₂	Methylene blue (10 mg/L)	High pressure mercury lamp (80 Watt)	60	95	-	Colpani et al. (2018)
TiO ₂ / β -cyclodextrin	4-nitrophenol (1.1×10^{-4} M)	Ultraviolet light	120	83 to 99.6	-	Subramanian and Ponnusamy (2017)
TiO ₂ /CeO ₂ / β -cyclodextrin	Rhodamine B	Sunlight	180	98.9	-	Gurunathan and Ponnusamy (2017)

method for eliminating cationic golden yellow X-GL/dye from wastewater. The prepared material degradation efficiency dropped from 80 to 50% at the end of the fourth cycle. The spent catalysts can be directly mixed with wall coatings and cement due to their non-toxicity.

Gum

Guar gum is a biodegradable polysaccharide produced by endosperms of guar beans composed of D-mannopyransoyl with D-galactopyransoyl units (Balachandramohan et al. 2020). Guar gum is employed as a supporting agent due to the ability to tune rheological properties. Guar gum can also act as a potential stabilizer for different catalysts. However, few studies have been reported guar gum/TiO₂ photocatalyst for wastewater treatment (Nasrollahzadeh et al. 2021). For instance, Rahimdokht et al. (2019) et al. employed TiO₂/gum tragacanth nano-hydrogel to redeem MB from textile wastewater. The catalyst exhibited 88.86% methylene blue removal at optimized pH 9.02, time 124.34 min, and an initial dye concentration of 9.37 mg/L.

Xanthan Gum is composed of repeating pentasaccharide units of two glucose units and two mannose units and one glucuronic acid. The structure also resembles cellulose. The important properties are non-toxicity, water-solubility, thermal stability, high viscosity. The Xanthan gum/TiO₂ was prepared through a simple mixing method followed by calcination. Experimental studies were conducted in the presence and absence of hydrogen peroxide (74%) and the studies under the influence of hydrogen peroxide exhibited greater catalytic activity. About 89% of methyl orange was removed in the presence of solar light. The higher adsorption ability and improved photo-response boosts-up the catalytic activity of the composite. The higher visible light absorption of the catalyst was confirmed through the narrowing the band gap to 2.8 eV (Inamuddin 2019).

Collagen

Similar to the biopolymers mentioned above, few studies are conducted on collagen-supported TiO₂ photocatalyst. Collagen is an abundant protein seen in mammals and solid wastes of tanning industries (Bazrafshan and Stylios 2019). The utilization of these collagens as catalyst support is highly effective and sustainable. Nagaraj et al. (2021) reported that the collagen nano-biosponge exhibited 95% degradation of rhodamine B under the illumination of natural sunlight. The bandgap of the prepared catalyst is 2.5 eV, whereas TiO₂ exhibits 3.35 eV. The amine functionalization on the TiO₂ surface is responsible for the bandgap reduction. Lower bandgap contributes to higher charge separation, extended lifetime electrons, and generated holes. Overall, these

materials act as a cost-effective and sustainable engineering materials for real life applications.

The N doped TiO_2 /Acid soluble collagen-polyvinyl pyrrolidone hybrid composite was developed by sol–gel method. The oxygen vacant sites and nitrogen interstitial type doping in N doped TiO_2 was confirmed with the x-ray photoelectron spectroscopy. Photocatalytic studies reported an 86% of naphthalene removal within 100 min under the visible light irradiation. The catalyst also extended the reusability for four cycles and the degradation efficiency is declined to 81.4%. The proposed degradation mechanism of naphthalene is reported in the Fig. 9. The possible mechanism of the photocatalyst is that the aromatic rings of the naphthalene is readily adsorbed on the surface of the photocatalyst due to the π - π stacking adsorption between the benzene rings of naphthalene and pyrrole rings of the photocatalyst. So, under the illumination of light, electrons reacts with oxygen to produce superoxide radicals. These superoxide radicals and holes plays a major role in elimination of pollutants (Doabi et al. 2022).

Industrial perspectives

The promotion of photocatalysis on an industrial scale is necessary. For that, the adaptations of structural changes of the photocatalyst are important to alter the band gap of photocatalyst and to utilize the visible light irradiation efficiently. The main problem that restricts photocatalysis at a larger scale is the recovery of the photocatalyst after each cycle. For example, the photocatalysts in powder form easily get precipitated at the bottom of the tank. So, the complete recovery of these photocatalysts is impossible. In addition to

this, the low absorbance of light and poor contact between powdered photocatalyst and pollutant. Thus, the adaptation of immobilization of the photocatalyst is the acceptable solution. As the aim of this manuscript, biopolymers can be employed as sustainable low-cost immobilizing agents. However, the complexity of the reactor design is a major problem. The need of maximizing the contact between the illuminated catalyst surface area and reactor volume reduces the path length of wastewater above the surface from a suitable light source. Despite these disadvantages, the pilot-scale studies using the immobilized catalysts were reported. The adaptation of biopolymers for the immobilized system also exhibits several advantages: biocompatibility, low cost, environmental friendliness and ease in preparation.

Conclusion

This review paper summarizes the utilization of different biopolymer-supported TiO_2 for the reclamation of organic pollutants seen in aquatic steams. The TiO_2 based photocatalyst is still considered one of the most prominent routes for the redemption of organic pollutants. Even after continuous research, TiO_2 suffers from several problems due to the aggregation of nano-sized TiO_2 , mass loss during catalyst recovery, and poor reusability. Numerous efforts have been put by the scientific community to address the challenges of TiO_2 as a photocatalyst. This study reviewed the different biopolymers as the supporting agents for TiO_2 . The natural polymers were selected due to their abundance, biodegradability, biocompatibility, non-toxicity. Several preparation routes are immobilization, hydrothermal, and sol–gel methods are well discussed.

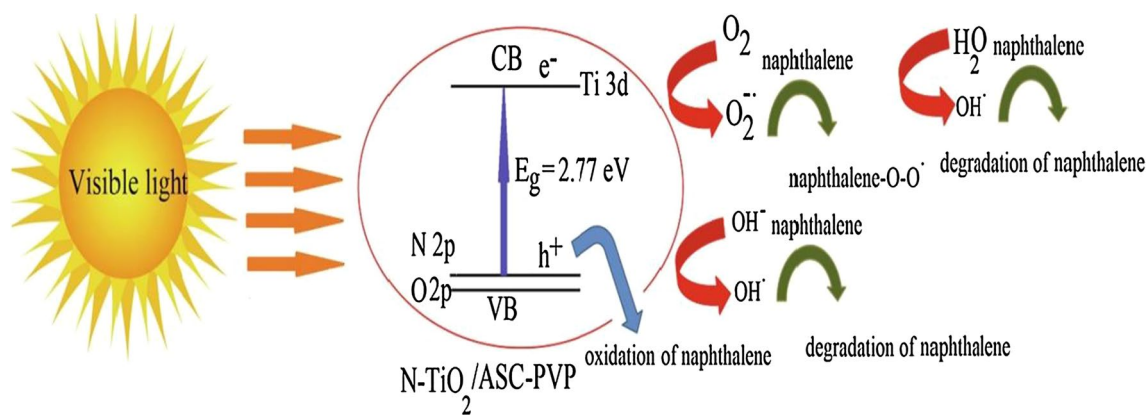


Fig. 9 Preparation mechanism of Nitrogen doped TiO_2 /Acid soluble collagen-polyvinyl pyrrolidone hybrid composite toward the removal of naphthalene. The illumination of visible light cause the electrons to move from valence band to conduction band. This leads to the simultaneous production of holes in the valence band and excess electrons in the conduction band. The holes hold a positive potential to

generate hydroxyl radicals and oxidize naphthalene. Reproduced from Doabi et al. (2022) with permission from Elsevier. Where N- TiO_2 /ASC-PVP is the nitrogen doped TiO_2 /acid soluble collagen-polyvinyl pyrrolidone composite, e is electrons, h is holes, CB is conduction band, VB is valence band and E_g is the band gap energy of the photocatalyst

The utilization of different biopolymers as a supporting medium to TiO₂ has gathered high efficiency and enhanced the reusability of the photocatalyst to larger extent. However, various challenges and further research recommendations must be addressed as earlier as possible to develop a sustainable wastewater treatment technology.

- The large-scale preparation of biopolymer-supported TiO₂ photocatalyst at minimum investments.
- Further research is mandatory in selecting suitable catalyst support based on selectivity and stability. Moreover, enhancing the active surface area to volume ratio and greater mass transfer is necessary for a photocatalytic process.
- Development of the metal-doped TiO₂ supported biopolymers to enhance the visible light activity of the photocatalyst. Moreover, metal doping is also beneficial in narrowing the bandgap to visible light. This may also increase the chances of complete utilization of natural sunlight as a light source.
- The development of magnetic TiO₂ supported biopolymer-based photocatalyst leads to a revolution in wastewater remediation, minimizing the problems related to catalyst loss and reusability.
- Application of different bio-waste materials as a renewable feedstocks for the development of biopolymers for wastewater remediation.
- The need of greener biological processes are necessary to bring down the production cost of biopolymer based materials under mild conditions and to improve the catalytic performance.
- In photocatalytic membranes, the impact of fouling is severe and leads to the deposition of organic pollutants on membrane surface that may deteriorate the degradation efficiency. Thus, the development of cost-effective fouling photocatalytic membranes is necessary to meet real-life applications.
- Moreover, the reduction in bandgap due to biopolymer incorporation needs further investigation. Overall, the role of biopolymer as a supporting agent for TiO₂ needs a detailed study to understand the mechanism in photo-degradation.
- Most current photocatalytic studies have only been conducted at the laboratory scale. The performance of the photocatalytic studies in the pilot plants will help us know the process feasibility.

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Declarations

Conflict of interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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