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Biological and Photocatalytic Degradation of Congo Red, a Diazo Sulfonated Substituted Dye: a Review

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Abstract The environment has undergone significant change because of technological advancement. Industries are releasing pollutants directly into the environment. Water pollution is a growing issue for humanity. Methods for managing wastewater generated by biological and industrial wastes are being developed. Textile industries are risking the health of living beings by contaminating water with dyes. Azo dyes are the major constituent of wastewater from textile industries. This review article focuses on the photodegradation of Congo red, the most prominent Azo dye. For Congo red degradation, both biological (via microorganisms) and chemical (via nanoparticles) methods are being investigated. The biological method primarily employs bacterial and fungal species. Bacterial species such as *Bacillus sp*., *Pseudomonas sp.*, and *Staphylococcus lentus* sp. efficiently degrade Congo red dye. The presence of functional groups on the cell wall of fungi, such as phosphates and hydroxyl, promotes efficient dye degradation. The use of nanoparticles for photodegradation of dyes is preferable because it does not result in polycyclic compounds after degradation. Many bimetallic catalysts, such as ZnO and TiO_2, have shown promising photocatalytic properties due to their large band gap. The use of nanoparticles that can be easily separated after photodegradation is preferred. As Gd^{3+} doped cobalt ferrite nanoparticles have higher removed capabilities than undoped cobalt ferrite nanoparticles, doping improves the degrading capability of nanocatalysts.

Keywords Azo dyes · Congo red · Photodegradation · Nanoparticles · Microbial degradation · Sulfonated dyes

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1 Introduction

The world's growing population is constantly interacting with and interfering with natural resources in order to improve living standards (Khan et al., 2018). Water is a valuable natural resource that is significantly affected by anthropogenic activities (Nandhini et al., 2019). No one can deny the importance of colors, so humans use a large number of colored substances, either natural pigments or manmade dyes, to add color to fabrics (Blaszczyk, 2019), food, leather, and paper. Natural substances are obtained from plants and algae. The use of natural pigments as colorants for dyeing on a commercial scale has been limited due to their less thermo-stability, photostability (Konieczkowska et al., 2015), difficulties in obtaining them from biota, and their less capability to remain fixed with the fabrics after periodic washing (Maddhinni et al., 2013; Simon et al., 2017). To fulfill

the requirement of these dyes, chemically synthesized dyes are common in the food, paper, and textile industries nowadays. Synthetic dyes are complex aromatic, poorly degraded colored compounds, mainly formed by using phenol and benzene (from petroleum) as starting materials (Mahapatra, 2016). There are different types of dyes; the most synthesized and used are azo dyes. Azo dyes contain –N = N– bonding and aromatics or heterocyclic compounds with recalcitrant structures (Khan et al., 2018; Tsuboy et al., 2007). The use of these compounds on a large scale and theoretical dumping of those color-loaded effluents without their degradation, into various natural compartments, antagonistically influences the harmony of these environmental frameworks (Khan et al., 2018). These dyes have the potential to absorb sunlight and reduce the photosynthetic activities of the aquatic flora (autotrophs) (Bianco Prevot et al., 2001). Their presence reduces the oxygen content in the water, thus causing the death of aerobes and ultimately disturbing the natural structure of freshwater bodies (Abe et al., 2017; Bianco Prevot et al., 2001; Zaini et al., 2014). Some of them are very toxic (Natarajan et al., 2011) and even cause a mutation in the DNA of aquatic life forms that leads to a malignant tumor formation in the brain (Fairuzi et al., 2018). Being genotoxic, mutagenic, and cancer-causing, these colors are very harmful to the soundness of biota (either oceanic or earthbound) (Kaushik & Malik, 2009b; Zaini et al., 2014). Textile industries are considered a big source of these contaminants as a huge amount of water used in the dying process. About 100 L of water is required for imparting color to 1 kg of pure cotton (Khan et al., 2016; Steingruber, 2000). A lot of these colors are impervious to debasement and remediation under normal conditions and through traditional strategies. These conditions have required the improvements of successful and effective wastewater treatment methodologies without disturbing nature and other living things (Krishnakumar et al., 2011; Rai et al., 2014). The removal of these water contaminants has gained much more attention over the last two decades. The purpose of this review is to assist researchers dealing with the degradation of Congo red (CR) dye by (i) assembling useful data on bacterial and fungal degradation; (ii) summarizing data on photodegradation of CR by nanoparticles (NPs); and (iii) discussing reaction mechanisms and intermediate products formed during degradation. Several reviews have been published that explain various methods for degrading dyes in textile wastewater. This review discusses the detailed mechanisms of biological degradation of CR, with a focus on bacterial and fungal degradation, as well as their shortcomings. A better method, photodegradation with nanocatalysts, has been thoroughly described, along with proposed mechanisms for efficiently removing CR from textile wastewater.

1.1 Origin of Dyes as Textile Colorants

Egyptians are considered the pioneers of fabric dyeing procedures. They are used to extract the colored pigments from naturally present entities. Chemical testing of several findings, for example, the red fabric from the ancient tomb of Egyptian King Tutankhamen, confirmed the existence of textile colorants (Yusuf et al., 2017). In the past, people used locally available colors for dyeing textile materials. These colorants were obtained from the plant's berries, fruits, leaves, roots, stem, or animal skin and minerals with little processing. But the applicability of natural pigments was limited on a commercial scale due to their less photo-stability, poor fixation with the material being colored, and fadedness over time. Thus, there arose a need to synthesize dyes artificially from petroleum products. William Henry Perkin (1856) discovered the first synthetic dye (Mauveine) in an unanticipated effort (Couto, 2009; Travis & Culture, 1990). This discovery urged scientists to synthesize dyes and test their applicability on a massive scale. Discovery of Mauveine also paved the path for immunologists and chemotherapists to test the effects of dyes on healthy as well as malfunctioned cells. Paul Ehrlich (1891) found that certain cells (mostly cancerous) took up a specific dose of dyes that started their death without causing damage to healthy cells (Parascandola, 1981). The reason for dyes to be bright in colors is their absorbance in the visible region because those compounds appeared colored which absorbed radiation in the visible region (400–800 nm). Colored compounds contain specific functional groups (chromophores, auxochromes) that absorb a particular photon of light with a characteristic wavelength and thus give a specific color to each compound. According to Witt's theory, auxochrome groups act as electron donors and play role in dyeing the materials while chromophore groups act as

electron acceptors. This theory was further worked on by two other scientists who stated that both the auxochromes and chromophores are linked through a conjugated system. They formulated a base for a new chromogen system (Fairuzi et al., 2018; Gürses et al., 2016). According to electronic structure theory, the colors of dyes are due to excitations of delocalized electrons (Schaefer III et al., 1995).

1.2 Recalcitrant Substances

Substances that possess complex structures, resistant to natural degradation, remain in the ecosystem for longer durations, and are transported from one geological location to another are termed as recalcitrant substances or pollutants (Singh et al., 2015). Most recalcitrant compounds in our environment are the results of oil, polymer, pharmaceutical, and textile industries (Zhang et al., 2019c). They are still present in our environment due to the inefficient treatment of wastewater treatment plants (Duan et al., 2018; Yarahmadi et al., 2018). Biological processes are still used for the removal of such substances from wastewater, but the process is relatively slower than modern technologies, such as photocatalysis, implied for wastewater treatment (Crini & Lichtfouse, 2019a; Zhang et al., 2018, 2019a, b).

1.2.1 Textile Dyes as Recalcitrant Substances

The molecular sizes of dyes are usually large due to the presence of bigger aromatic rings and heavy substitutions; as a result, dyes resist natural degradation and pose serious harm to the environment (Kaushik & Malik, 2009b). In industries, structures of synthetic dyes are modified by incorporating certain biological, chemical, and physical agents. This enhances their photo-stability as well as their ability to resist fading over a long time (Shabir et al., 2017). The presence of these agents makes dyes defiant towards degradation. Synthetic dyes containing chromophores and auxochromes are highly reactive. When discharged into a natural environment, they react with several pollutants, for example, heavy metals and halogens, and form even more carcinogenic substances. Recalcitrant substances contribute significantly to the reduction of oxygen in water bodies, resulting in eutrophication; as a result, the availability of oxygen to aquatic biota decreases, limiting photosynthetic activities in these bodies and disrupting the food chain. (Jorfi et al., 2018; Kaushik & Malik, 2009b). Moreover, when such toxic entities enter the living bodies, they adversely affect the DNA of the cells as they tend to bind with the proteins. Many dyes have been proved to cause severe diseases such as cancer (from azo dyes). Recalcitrant substances equally affect aquatic and plant life. The synthetic color from the dyes also inhibits the natural photosynthetic procedure of aquatic plants (Mohapatra et al., 2020). For the removal of pollutants from the environment, various methods are being used over the past few years. Advanced oxidation processes (AOPs) used ozone gas along with hydrogen peroxide or ultraviolet light, H_2O_2/ultraviolet radiation, the photo-Fenton process, and the Fenton process for the removal of recalcitrant structures or to convert them into less harmful more biodegradable substances (Feuzer-Matos et al., 2021; Tahir et al., 2016). The most common dye present in the wastewaters is an azo dye.

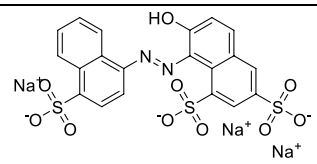
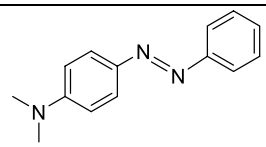
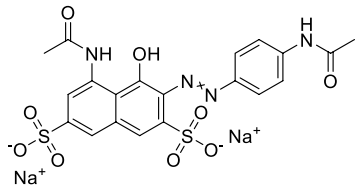
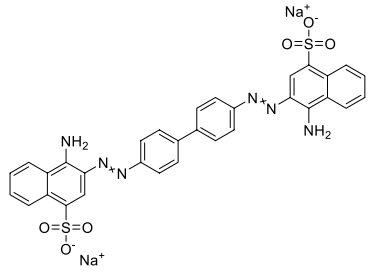
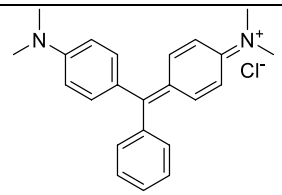
1.2.2 Azo Dyes

Almost 9 lac tons of dyes are discharged into the aquatic environment annually, of which 70% are azo dyes (Balapure et al., 2015; Selvaraj et al., 2021). Azo dyes have been identified as known carcinogens for the human urinary bladder (Sen et al., 2016a). They are also known to cause liver and spleen cancers in humans (Solís et al., 2012a). More than 3000 azo dyes are being manufactured annually all over the world. Like many other dyes, azo dyes are also responsible to change the natural factors of water bodies such as color, oxygen demands, and pH. (Selvaraj et al., 2021). Common azo dyes and their applications are described below in Table 1.

1.2.3 Congo Red (CR)

Congo red (CR) is an important azo dye as it is used for dyeing of the cotton fabrics. They are known carcinogens (Selvaraj et al., 2021). The CR is an important sulfonated substituted benzidine diazo dye (Olivo-Alanis et al., 2018), which is considered a more recalcitrant compound than other mono and diazo dyes present in untreated wastewater. It can be used in leather, paper, plastic, printing, and textile

Table 1 Common azo dyes and their hazardous effects

Dye	Structure	Applications	Effects	References
Brilliant Red		Cosmetics, drugs, food, and textile industries.	Muscle spasm.	(Gao et al., 2018)
Red Dye		Textile industries.	Malignant tumors in humans.	(Puvaneswari et al., 2006)
Acid Violet 7		Food, paper, and textile industries.	Chromosomal aberration.	(Moosvi et al., 2005)
Congo Red		Textile industries and as a pH indicator.	Carcinogen.	(Sudha et al., 2014)
Malachite Green		Leather and paper industries.	Carcinogen and mutagen.	(Khehra et al., 2005)

industries as a coloring agent. Furthermore, its use as a pH indicator for the diagnosis of amyloidosis (Khurana et al., 2001; Puchtler et al., 1962) and in the detection of free HCl (hydrochloric acid) in gastric juice was reported (Shu et al., 2015). Due to its resistance to natural degradation, it causes spoilation of water bodies and the landmass (Dubé et al., 2008). UV–visible spectra of CR showed three distinct bands (Fig. 1) at 498 nm, 347 nm, and 235 nm attributed to the chromophore group, naphthalene, and benzoic ring respectively (Movahedi et al., 2009a). Table 2 describes some important features of CR.

2 Methods for the Degradation of Dyes

The removal of colored substances from the wastewater by conventional methods, such as adsorption (Banerjee et al., 2019), AOPs (Kanakaraju et al., 2018; Kraft et al., 2003; Tröster et al., 2002), coagulation (Hamoud et al., 2017; Sohrabi et al., 2017),

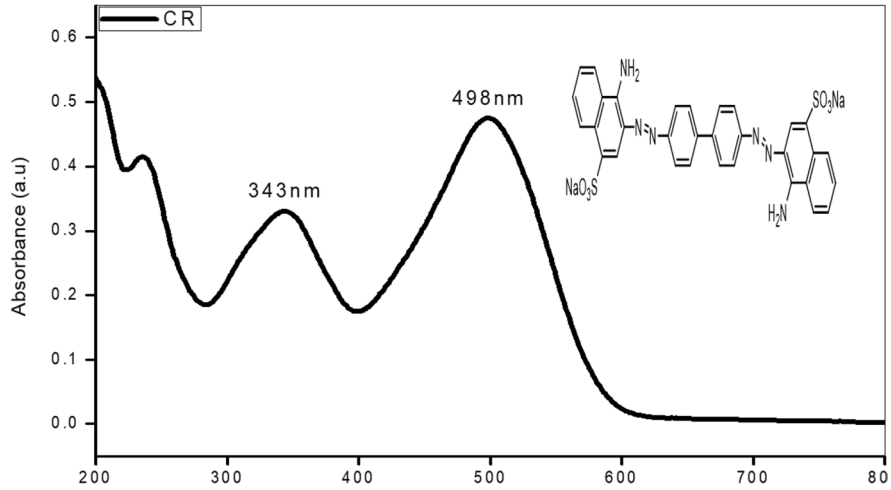


Fig. 1 UV–visible spectrum of CR

Table 2 Characteristics of CR

Parameter	Remark
Dye	Congo red
Formula	$C_{32}H_{22}N_6Na_2O_6S_2$
Lambda max	498 nm, 343 nm
Molecular weight	696.665gmol^{-1}
IUPAC name	3,3′-([1,1′-biphenyl]-4,4′-diyl)bis(4-aminonaphthalene-1-sulfonic acid)
CAS number	573–58-0
ChemSpider	10,838
pH	3–5.2
Classification	Diazo sulfonated substituted organic compound
Nature	Recalcitrant substance

Fenton process (Cruz-Rizo et al., 2017; Sohrabi et al., 2017), flocculation (Beluci et al., 2019; Sohrabi et al., 2017), ozonation (Mahmoodi, 2016; Manivel et al., 2015; Tapalad et al., 2008; Wu et al., 2015), and ultra-filtration (Korenak et al., 2018) (as shown in Fig. 2), was common in the last few decades, but all these methods were proved insufficient for the complete removal or transformation of toxic substances into eco-friendly substances because of high expense and high energy requirements (Chao et al., 2018). The application of such methods is thus prohibited due to additional energy requirements, high costs as compared to modern processes (Ekambaram et al., 2018), problems associated with the operation, production of

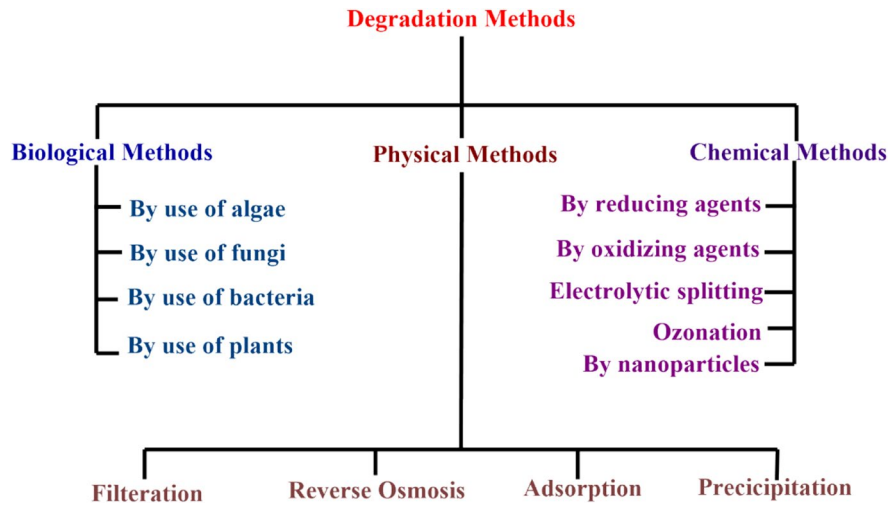


Fig. 2 Different methods for the degradation of dyes

a lot of sludge, and unsafe disposal, which can act as a secondary pollutant.

Many researchers have explored different methods, bioremediation (Mahmoud et al., 2017), phytoremediation (Ekambaram et al., 2018; Saraswathi et al., 2017), and degradation by using NPs (Lamba et al., 2015; Mahdiani et al., 2018; Qu et al., 2018) for the manipulation of dye-laden (Fig. 3). All these methods have their pros and cons.

2.1 Biological Methods for the Degradation of Dyes

AOPs require additional energy and space, high costs, and advanced instruments, for the treatment of wastewater pollutants as compared to photocatalytic degradation. They are also less efficient in terms of generating huge quantities of sludge, and the transformation of colorants into more harmful secondary pollutants (Pandey et al., 2007). The AOP has yet been successful in the past years in the mitigation of harmful impacts of dyes. AOPs bring physical changes in the chemical structure of pollutants (Alderete et al., 2021). On the other hand, biological methods are considered environment-friendly as they perform complete or nearly complete degradation of toxic effluents (Solís et al., 2012b). Microorganisms possessed the metabolic potential for the degradation of pollutants or xenobiotic compounds, due to the diversity of lysosomal enzymes (approximately 40 different types of enzymes, such as acid phosphatases, lipases, sulfatases). These enzymes are present in many species of algae, bacteria, fungi, and plants. Microbial decoloration by bacteria is considered an efficient method (Nigam et al., 1996). Bacteria that degrade synthetic pigments can easily be obtained from animal and human excreta, food, soil, and water contaminated with microbes. The degradation of dyes by using living organisms is a vital research area in environmental biology. The success of biological methods in treating effluents is basically due to the ability of biota to acclimatize themselves for toxic substances and development of resistance against these substances; as a result, they utilized toxic compounds and convert them into the more stable, less toxic form (Carliell et al., 1996). In the microbial degradation of dyes, there are two ways to convert dyes into less toxic substances: first is the adsorption of dyes onto the biomass of microbes, and second is the degradation of dyes intracellularly (Carliell et al., 1996). The fungal degradation of pollutants is associated with the extensive extracellular enzyme system (Lai et al., 2017). Among different cultures of fungi, white-rot fungi are considered best for degradation. Many species of marine algae were also investigated for their abilities to degrade pollutants. Phytoremediation is also another technique to handle pollutants (Saraswathi et al., 2017). Both of these techniques have their advantages as well as limitations; however, all these work efficiently only at

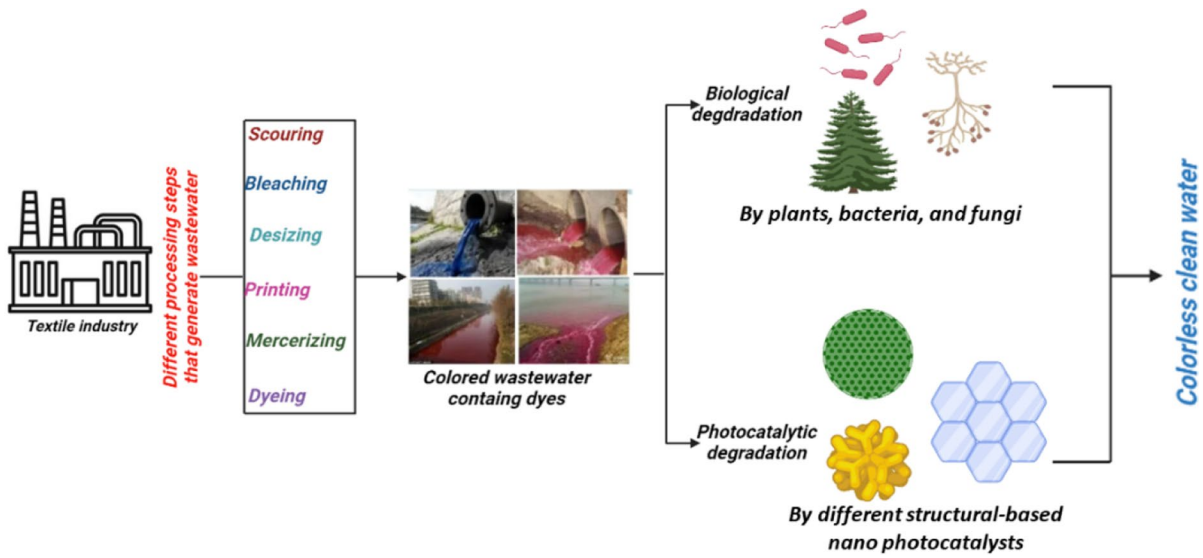


Fig. 3 Production of wastewater from the textile industry and its decolorization

optimum conditions like concentration of dye, energy sources for microbes, humidity, pH, structure of dye, temperature, and oxygen content available to biota (specific in discoloration of dyes) (Singh et al., 2012).

2.1.1 *Bacterial Degradation*

Our environment is largely dependent upon the nutrient cycles taking place in our surroundings involving bacteria at various steps. Still, several areas, which may prove helpful in innovative ways, are yet to be explored or are on the verge of recognition by scientists (Telke et al., 2010b). Bacteria initiate the degradation of dyes by cleaving azo bond/s either in the presence or absence of oxygen due to the action of their enzymes as shown in Fig. 4. The culture of *Bacillus subtilis* was first isolated and reported as the dye decolorizing in the 1970s (Horitsu et al., 1977). Isolation of dye decolorizing cultures of bacteria seemed to be a difficult task. Many aerobic and anaerobic bacterial strains have been found to decolorize the azo and other nitrogenous dyes efficiently. Azoreductases and laccases are two major families of enzymes present in all these strains that possessed the potential to metabolize many industrial dyes. Azoreductases need NADPH, FADH, and NADH as reducing equivalents to reduce the dyes completely into colorless amines (Sarkar et al., 2017). Many species of bacteria, yeast, filamentous fungi, and plants have been reported that contain genes in their DNA for coding oxidative enzymes like lignin peroxidase and manganese peroxidase, capable to degrade dyes. Fungal cultures have proved their rapid efficiency in the degradation of dyes because they possess many ligninolytic enzymes. Fungi may perform the decoration by the adsorption process or enzymatic degradation or it may occur as a collective procedure of both (Sen et al., 2016b). Instead of individual bacterial cultures which are rather time-consuming to extract, mixed bacterial cultures are preferred but the results are not as promising as they would be with the individual cultures of aerobic, anaerobic, and anoxic bacterial conditions (Jamee et al., 2019).

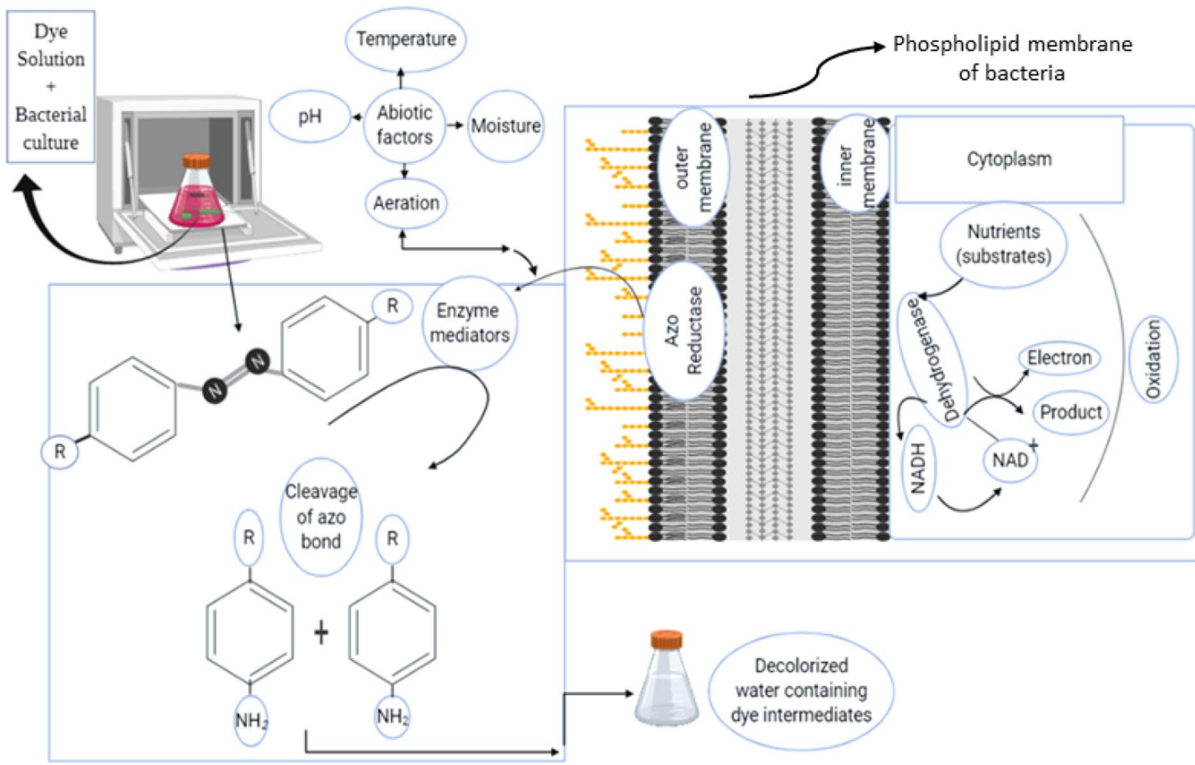


Fig. 4 Degradation of dyes by bacteria

2.1.2 Fungal Degradation

Fungi have also been used for biodegradation purposes. Their application in the decolorization of wastewater is numerous (Blais, 2006; Kaushik & Malik, 2009a). Fungi tend to degrade textile dyes by secreting extracellular enzymes oxidases and reductases as shown in Fig. 5. This is a factor that provides an advantage of fungal degradation over bacterial degradation. The large size of fungi as compared to bacteria also favors fungal degradation (Oliveira et al., 2007). Sometimes, enzymes are isolated from the fungi and used independently (Cristóvão et al., 2009). Moreover, fungi are capable of adapting themselves according to the environment unlike most bacteria (Ahmad et al., 2015; Rahimnejad et al., 2015). Some of the most important factors that control the degradation of textile dyes via fungi include temperature, pH, pretreatment, and the presence of some nutrient compounds (Arıca & Bayramoğlu, 2007; Kapdan & Kargi, 2002; Neoh et al., 2015). Spores of fungi have also been reported to remove pollutants from water (Hoque & Fritscher, 2019; Li et al., 2019).

2.2 Photodegradation

Photocatalytic degradation of dyes has several advantages over other methods. They involve the use of heterogeneous semiconductors that upon absorption of light degrade the toxic dyes (Rafiq et al., 2021). The photodegradation of dyes is controlled by several parameters including dye concentration, pH, and temperature (Ikram et al., 2020). Comparative to biological and other conventional methods which require a certain type of pretreatments to be done before treating organic pollutants (Fernández et al., 2010), photocatalysis may be a better choice. Photocatalytic degradation involving heterogeneous catalysts proves to be an efficient

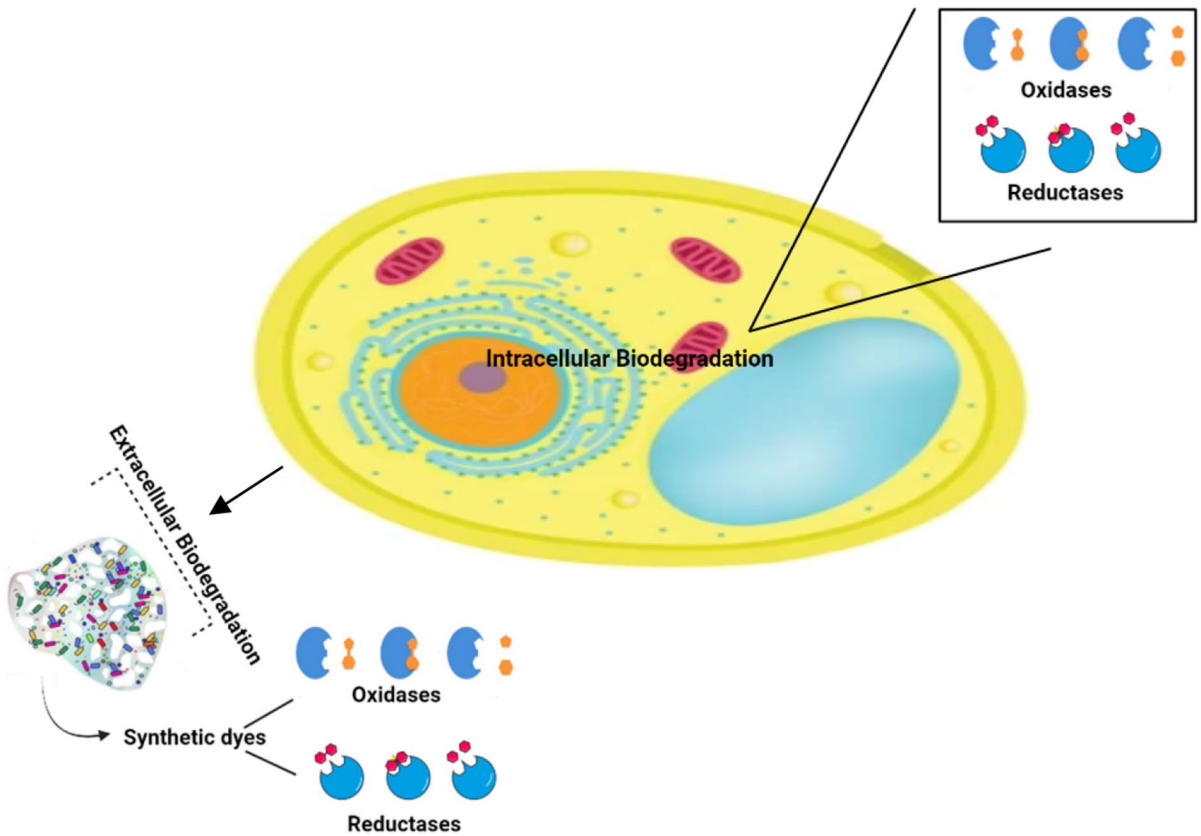


Fig. 5 Fungal degradation of textile dyes

way to treat persistent organic pollutants (Singh et al., 2016). Photocatalysis possesses the ability to degrade poisonous organic pollutant into their least harmful form for example carbon dioxide using some sort of nanocatalyst (Meephon et al., 2019); photocatalysts on absorbing UV or visible light (depending upon their nature) generate high energy photons which further reduce the organic pollutants (Aslam et al., 2022). Photocatalysts generally involve semiconductor catalysts; they may be monometallic (consisting of one atom), bimetallic (consisting of two atoms), or tri-metallic (consisting of three atoms) (Rajendran et al., 2021a). To improve the efficiency of a photocatalyst, charge carriers are increased. The more the charge carriers are, the more efficient is the degradation. To improve the degradation capacity, semiconductors are often doped with other metals (Terna et al., 2021a). To understand the principle of photodegradation, we first need to understand the structure of a semiconductor. Semiconductor consists of two bands: a conduction band (CB) and a valence band (VB). There is a gap between these two bands, as shown in Fig. 6, called the band gap. When electrons present in the VB absorb energy equal to or greater than the band gap, they are promoted to the CB generating a hole (h^+) in the VB and an electron (e^-) in CB. These two, a hole (h^+) and an excited electron (e^-), are the main cause behind photocatalysis. Photogenerated e^- in the CB and h^+ in the VB act as oxidizing and reducing agents respectively (Bhat et al., 2020a; Pattnaik et al., 2018a; Vidya et al., 2017a). The efficiency of photocatalysts can be improved by various methods including doping and metallization. They tend to increase the charge separation of photocatalysts (Arkhipov et al., 2005a).

3 Degradation of Congo Red by Bacteria

Microorganisms could be acclimatized by providing different nutrients initially, and then exposing them to different concentrations of dyes (ranging from low to high). As soon as microorganisms become acclimatized, they start using dyes as their energy source as a result of recalcitrant substances degraded (Gopinath et al., 2009). The degradation of dyes by bacterial culture in the presence of sufficient nutrients could produce colorless intermediates and final products which may prove even more toxic than parent compounds. Thus, decoloration of dyes cannot be considered a single parameter to interpret percentage degradation. Proposed mechanism of biodegradation of CR is shown in Fig. 7.

K. P. Gopinath et al. reported nutrient-free degradation of CR by a *Bacillus* sp. Dye samples were

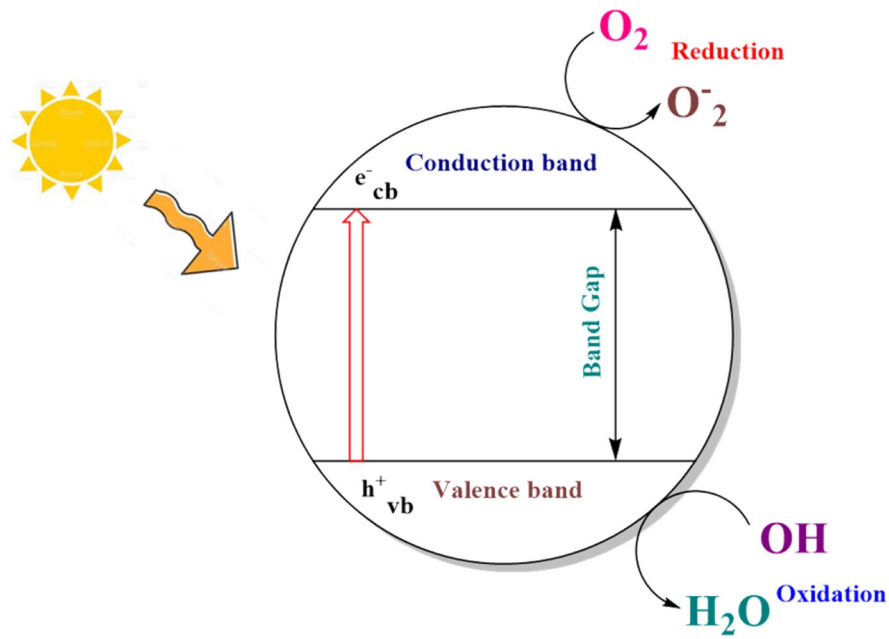


Fig. 6 General mechanism of photodegradation

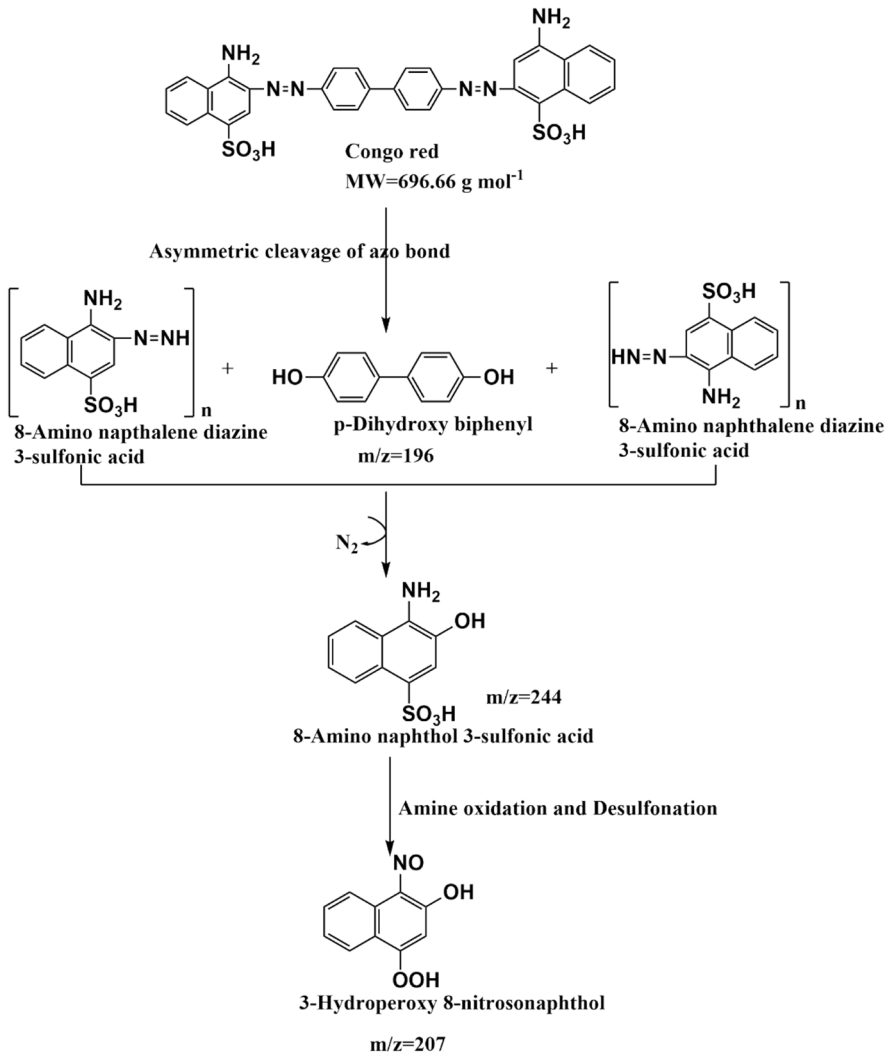


Fig. 7 The proposed mechanism of degradation of CR by microbes (Telke et al., 2010a)

pretreated with sonication and results were compared with non-pretreated samples. Sonication of dye samples with low-energy ultrasound created oxidants by the homolytic division of bonds present in water containing dye. *Bacillus* sp. decolorized a non-pretreated 100–300 mgL^{-1} sample of CR within 24–27 h of incubation, giving 100% degradation efficacy, 97% reduction in COD, at 7 pH, and 37 °C temperature. While with pretreated samples the incubation period was reduced to 6–7 h, in such case, the other parameters remained the same (Gopinath et al., 2009). One of the important findings by Amar et al. showed 97% degradation of 100 mgL^{-1} CR by a bacterium *Pseudomonas* sp. Su-EBT in 12 h with a 50% decrease in COD at 8 pH and 40 °C temperature (Telke et al., 2010b). It was reported that the degradation rate is maximum in static conditions although the bacterial growth rate is more in shaking conditions (Telke et al., 2009). The slow rate with less degradation efficacy in shaking conditions was attributed to the suppressed activity of laccase (one of the oxidases responsible for degradation). The time for degradation increased as the concentration of CR dye increased and the maximum degradation of CR is 97% within 60 h at a $1gL^{-1}$ concentration (the maximum tolerable amount of CR by *Pseudomonas* sp. Su-EBT) (Telke et al., 2010b). Dyes with fewer sulfonate groups decolorized rapidly than dyes with more sulfonate groups (Pearce et al., 2003). The degradation of dyes by bacteria is attributed to the adsorption of dyes to microbial cells. The degradation of CR has been described by

Kamel Cheib et al. using a bacterium, *Staphylococcus lentus*. *S. lentus* decolorized a 100mgL^{-1} of CR (97.3% degradation efficacy) when incubated for 24 h, in the presence of medium containing mineral salts, yeast extract 0.10% w/v, and glucose (7 mM) at 7.2 pH and 37 °C temperature (Chaieb et al., 2016). The inoculum size used 2.2×10^{6} colony-forming units mL^{-1} of liquid, in shaking conditions, contradictory to the results reported by Amar et al. The FT-IR spectrum of degraded CR confirmed the degradation as the position of peaks was found different than the non-degraded solution of CR. Moreover, phototoxicity test showed that the non-degraded solution of CR inhibits the germination of plumule and radicle of *Triticum aestivum* and sorghum bicolor, while degraded CR increases germination efficacy (Chaieb et al., 2016). The degradation of dyes in nutrient-rich medium containing microbial cells in static as well as shaking conditions results in loss of biomass (Bedekar et al., 2014; Girish, 2019). Alternatively, bioreactors (continuous or batch) are used for the degradation of dyes in which either a pure culture of bacteria or consortia of bacteria is used. Single cultures of bacteria mineralized only a few dyes specific to them, but are unable to mineralize higher molecular weight substituted, versatile dyes (Bedekar et al., 2014). In addition, consortia of bacteria were found more resistant towards diverse changing conditions and parameters such as concentration of dyes, pH, and temperature (Jalandoni-Buan et al., 2015). In such cases, degraded products of a dye by one kind of bacteria might act as nutrients for other kinds of bacteria, resulting in a maximum reduction in toxicity (Lade et al., 2015). In bioreactors, microbial cultures are immobilized on a carrier such as polyurethane, which efficiently holds the culture fixed and enhances the chances to reuse the same culture (Lade et al., 2015). A biocatalyst laccase has shown some promising results in wastewater treatment (mechanism shown in Fig. 8). But it only works if it has been immobilized and sustained its pH and thermal stability. Georgieva et al.

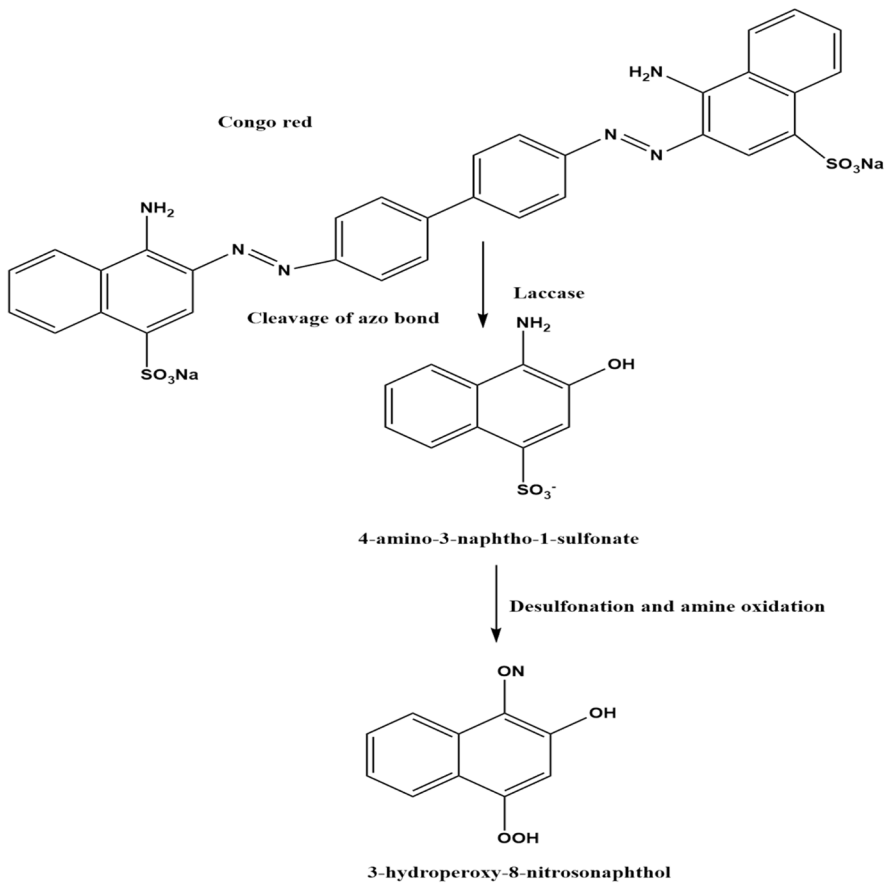


Fig. 8 The proposed mechanism for the degradation of CR by laccase of *Dietzia* sp. (Babu et al., 2015a)

Table 3 Different conditions for the biodegradation of CR

Bacteria	Conditions pH Temperature (°C) Substrates	Conc. of dye (mgL^{-1})	Inoculum	Incubation time (h)	% decoloration % COD removal	References
Bacillus sp.	Aerobic, shaking 7 37	100–300 Dye itself	1%V/V	24–27 6–7	93 - 100 97	Gopinath et al., 2009)
Pseudomonas sp. Su-EBT	Aerobic, static 8 40 Glucose	100 $1gL^{-1}$	50 CFU/mL	12 60	90 50 97 50	Telke et al., 2010b)
Staphylococcus lentus	- Shaking 7.2 37	100 0.10% w/v yeast extract, 7 mM glucose, mineral salts	2.2×10^6 CFU/mL	24	97.3 -	Chaieb et al., 2016)
Lysinibacillus sphaericus JD1103	- Shaking 6 28	- 1,2-Dihydroxybenzene, 3,4-dimethylbenzyl alcohol	-	72	84 ± 0.50 -	Chantarasiri et al., 2017)
Dietzia sp. (DTS26)	- Static $8, 32\pm2$	100 –	1.2×10^6 CFU/mL	30	94.5 86.4	Babu et al., 2015b)
Enterococcus faecalis L2C	- - -	700 1000 0.1% glucose 1% yeast extract	0.1 CFU/mL	168	97.7 - 82.6 -	Santoso et al., 2016)
Lysobacter sp. *P28*	Aerobic Static 7–8 37	50 – 200 –	1%V/V	24 72 120	73.5 - 84.4 - 58.3 -	Ranga et al., 2015)
Pseudomonas stutzeri L1	Anaerobic–Aerobic - 8 37	100 2% starch 0.2% yeast extract	23×10^4 CFU/mL	96	84 -	Kuppusamy et al., 2017)
Consortia of *P. stutzeri* and *Acinetobacter*	Aerobic - 8 37	100 2% starch 0.2% yeast extract	$23\times10^4+23\times10^4$ CFU/mL	120	97 -	Kuppusamy et al., 2017)

reported that immobilization of laccase over the membrane of polypropylene enhanced the catalytic performance to a great extent (Zhou et al., 2021). Table 3 describes the biodegradation of CR by different bacteria at different conditions.

4 The Degradation of Congo Red by Fungi

Due to the extensive and efficient extracellular enzyme system of fungal culture, mycoremediation of industrial wastewater has received a lot of attention (Pundalik et al., (n.d); Noman et al., 2020). Different fungal species and their degradation efficiencies have been shown in Table 4. Dye degradation by fungal mycelia is associated with the adsorption of dye to the cell wall of fungi (Chakraborty et al., 2013). Functional groups present in the cell walls of fungi help the dyes to bind through electrostatic forces of interactions (Skanda et al., 2021). Reactive functional groups such as carboxyl, hydroxyl, and phosphate are present in hetero-polysaccharide and a hydrophilic lipid portion of the fungal cell wall is responsible for attractive forces, leading to biosorption of dye solution with the cell wall; thus, the color intensity of dye is reduced, indicating conversion of dye into less toxic colorless metabolites (Chakraborty

Table 4 The degradation of CR by fungi and comparison of % degradation efficiencies

Fungus used	Conc. of dye (mgL^{-1}) Substrates	Condition Agitation speed (rpm) pH, Temperature (°C)	Phenomena Incubation time (h)	% degradation	References
Alternaria alternata CMERI F6	600 Yeast extract, glucose	Aeration 150 5 25	In vivo biodegradation 48	99.99	Chakraborty et al., 2013)
Aspergillus niger	200 Potato dextrose	Aeration 120–150 5 28	In vivo biodegradation 144	97	Asses et al., 2018)
Oudemansiella canarii	50 Sugarcane bagasse-wheat bran	Aeration 100 5.5, 30	In vitro biodegradation 24	80	Iark et al., 2019)
Pleurotus ostreatus MTCC 142	- Paddy straw and corn husk	Aeration - 3, 35	In vitro biodegradation 1	82.8	Das et al., 2016)
Ceriporia lacerata ZJSY	$0.1mgmL^{-1}$ IM and potato dextrose	Open-air Static 8, 35	In vitro biodegradation 48	90	Wang et al., 2017)
Pleurotus sajor-caju	150 Banana peel extract	Aeration 4, 35	In vitro biodegradation 1	95	Yehia et al., 2017)

et al., 2013; Singh et al., 2020). In mycoremediation of industrial effluents, several parameters like aeration, agitation speed, fungal species used, initial dye concentration, incubation time, nutrition sources, pH, presence of heavy metals, and temperature all affect degradation rates. Different substrates have been used by many researchers, to check the effect of nutrition sources on the degradation rate. Inorganic media containing glucose were reported as the best carbon source for fungal growth and maximum decoloration efficacy of CR. Shaking speed plays an important role in controlling the degradation rate (Asses et al., 2018). The degradation of azo dyes is greatly enhanced if the nutrient substrate contains carbon and nitrogen. Carbon and nitrogen act as the donors of electrons which are then accepted by the azo dyes which help in the degradation of dyes by the generation of reducing equivalents (Kapoor et al., 2021). Shaking promotes the growth of fungal culture by increasing the mixing of oxygen with biomass; as a result, degradation efficacy of dyes increases. Shaking speed from 0 to 250 rpm was reported by different authors. *Aspergillus* sp. showed the maximum degradation rate at an optimum shaking speed of 150 rpm for the degradation of CR; it was attributed to the maximum enzymatic activity of manganese peroxidase (MnP) and lignin peroxidase (LiP) (Asses et al., 2018). Optimum temperature is another factor that contributes to degradation. It was reported that extracellular enzymes of fungi like MnP and LiP showed maximum activities in temperature ranges between 25 and 35 °C (Parshetti et al., 2007). Enzymes are sensitive to high and low temperatures resulting in denaturation and inactivation, respectively. As the initial concentration of dye increased, the rate of degradation decreased; this could be attributed to the saturation of active sites of enzymes. Catalytic proteins are found sensitive to pH; thus, most of them work in a narrow range of pH (5–8) due to their structural stability in acidic conditions (Iark et al., 2019).

5 Biological Methods and Their Drawbacks

Microorganisms and adsorption/sorption techniques are the most commonly used biological methods for dye removal (Bal & Thakur, 2021). Although biological methods are often regarded as environmentally friendly, they did not always meet our expectations when it came to wastewater treatment. The use of enzymes, for example, is not always successful because the process requires optimal conditions to be met and they do not completely remove the dyes from wastewater. Even minor changes in the conditions or parameters

result in a complete loss of enzyme activity (Bento et al., 2020; Carolin et al., 2021). Biodegradation is also sensitive to certain experimental conditions such as accumulation of ammonia inside the reactor (Garcia et al., 2020). Biosorption also causes biomass accumulation, which prevents the optimum process from establishing (M-Ridha et al. 2020; Sosa-Martínez et al., 2020). Another factor that contributes towards the ineffectiveness of biological and other conventional treatment methodologies is the chemical instability in the different organic mediums (Ali, 2010). When azo dyes were treated with bacterial cultures, they frequently produced toxic amines, resulting in secondary pollution (Brüschweiler & Merlot, 2017). Table 5 describes some advantages and disadvantages of photodegradation and biodegradation.

6 Nanoparticles as a Preferred Mode for Degrading

Nano-photocatalysts have long been used in wastewater treatment. Several nanocatalysts have demonstrated outstanding performance in degrading textile dyes from water. The most important feature of nanomaterials is their use of energy-efficient pathways. Furthermore, several structural changes enable greater light absorption and less recombination (Molinari et al., 2020). According to Ch-Th et al., 2021, the techniques used for the modifications are nanoscale designing and doping (Ch-Th et al., 2021). Tofa et al., 2019) and Uheida et al., 2021) degraded LDPE and microplastics using zinc nanorods with improved visible light photocatalytic efficiency modified from ZnO seed layers (Tofa et al., 2019; Uheida et al., 2021). Several studies show that nano-photocatalysts are more efficient than biological methods. Green synthesis allows nanoparticles to be synthesized with less environmental impact and provides nanoparticles with unique properties such as nontoxicity, high stability, and cost effectiveness (Chauhan et al., 2020). Biosynthesized ZnO nanoparticles demonstrated excellent photodegradation activities towards dyes such as CR (Weldegebrieal, 2020) and other organic compounds (Chauhan et al., 2020). Furthermore, we can summarize the advantages of photodegradation as follows (Khan et al., 2020b).

- Use of a nanocatalyst limits the need of pre- and post-treatments of the organic compounds to be degraded.
- Nanocatalysts are liable to recovery and can be reused.
- High chemical stability in various organic environments.
- No external arrangements are required to carry on photodegradation such as temperature and pressure.
- Variety of organic compounds can be degraded by nanocatalysts.

7 Photodegradation of Congo Red by Nanoparticles

One of the important green chemistry technologies is the photodegradation of organic dyes. Photocatalysts (PCs) are required for the degradation of dye. In photocatalysis, dyes are degraded under the influence of UV–visible radiations. This method is beneficial as compared to the conventional methods of pigment degradation because it produces no polycyclic products (Nagajyothi et al., 2020). Photodegradation by nanoparticles (NPs) bears many advantages; the most important is the complete degradation of dyes (Banerjee et al., 2019). Due to the active sites, catalytic potential, high surface area, high chemical stability, low cost, nontoxicity, and small size (Gunjakar et al., 2011), nanomaterials have proved themselves as efficient adsorbents (Raval et al., 2016). Photocatalytic degradation happens by using heterogeneous catalysts such as ZnO, Fe_2O_3, TiO_2, CdS, and ZnS under the influence of solar energy (Merah, 2020). ZnO NPs exhibit worthwhile properties regarding the degradation of organic dyes due to their wide band gap ranges and act as a promising semiconductor in photocatalytic activity (Nguyen et al., 2021). ZnO semiconductor falls under the category of N-type, and when it is irradiated with solar light and UV radiations, it produces electrons and holes which proves to be very helpful in the degradation of dyes completely (Debnath et al., 2020). NiO NPs are extremely efficient in dye degradation activity because of the super antioxidant properties that they are going to be used to provide clean water for drinking purposes without any pollutants and dyes present in the industrial wastewater (Bhat et al., 2020b). The concentration of the catalyst NiO has directly affected the antioxidant

Table 5 Comparison of biodegradation and photodegradation of dyes

Key factors	Biodegradation	Photodegradation	References
Time requirement	*Biological processes require a longer period* -It takes a lot of time to prepare a fungal culture for biotreatment -Some lignin cultures require 4–8 weeks prior biodegrading activity	-*Nanoparticles* Photodegradation of 50% methylene blue by SnO catalyst was completed in 47 min -*Colloidal NPs* in wastewater treatment require time in order of few seconds	Zabed et al., 2018), Niculescu et al., 2021), El-Salamony et al., 2018), Zabed et al., 2019)
Secondary pollution	-*Harmful waste production* Different biological treatments produce secondary pollutants that are highly toxic, e.g., leachate treatments transfer NH^{4+} in other environmental media	-Photodegradation does not produce toxic waste -Carbon-based nanoparticles (CNPs) which are used in photocatalytic activities produce less toxic secondary pollutants and follow green routes	Sajid & Płotka-Wasylka, 2020), Chen et al., 2022), Rafiq et al., 2021)
Cost effectiveness	-Conventional biological methods are expensive and time-consuming For the removal of heavy metal (e.g., arsenic, cadmium, and mercury), these methods prove to be less efficient and expensive -Even if *active bioreactors* are made, it is very expensive to achieve their full functionality	-*Nanotechnology is cost effective* Iron-based NPs are used in heavy metal removal from wastewater with less cost and effective removal -Use of *semiconductors* as nanocatalysts provides a cost-effective route for the photodegradation of organic compounds. For instance. BiOCl is used for the photodegradation of antibiotics	Hazarika et al., 2022), Khan et al., 2021a), Singh et al., 2022), Rambabu et al., 2020), HE et al., 2021)
Preparation and requirements	-*Biological methods* including aerobic lagoon (AL) and anaerobic filters (AF) are usually used in wastewater treatment processes. These methods require very high temperature, high retention time (HRT), and at least 5–11 days. The efficiency of procedure mainly depends upon availability of land in urban areas -*Selection of microbes* is also a great task for the biodegradation process. Moreover, sometimes, microbial consortia are required for efficient degradation which makes it difficult for the individual microbes to work at their optimum conditions	-*Nanoparticles* are usually easy to prepare and easy to handle. Routes for the preparation of NPs are eco-friendly and do not require huge land area. Nanotechnology is not vulnerable to climate change and can resist certain temperature changes as well -*Alternative light sources* also provide an efficient access for the photodegradation	Musa & Idrus, 2021), Parveen et al., 2016), Saravanan et al., 2021), Domínguez-Cuevas et al., 2006), Singh & Borthakur, 2018), Varshney et al., 2016)
Stability and biocompatibility	*Use of conventional biological methods* -Their preparation methods are time taking and produce secondary toxins. They are also non-biocompatible	*Magnetic nanoparticles* (MNPs) are highly stable and biocompatible, e.g., silica-coated MNPs	Li et al., 2021), Mohamed et al., 2022), Aithal & Aithal, 2022), Daniel et al., 2022)

Table 5 (continued)

Key factors	Biodegradation	Photodegradation	References
Biodegradability	-Biological methods including *bioreactors*, *microbiological treatments*, and *biological active sludge* produce non-biodegradable sludges that are very difficult to treat. These conventional methods have poor biodegradability	-*Superparamagnetic iron oxide* (SPION) NPs have superparamagnetic and paramagnetic properties and are significantly biodegradable -*Poly-D-L-lactide-co-glycolide* (PLGA) nanoparticles have nontoxic and biodegradable by-products -*Chitosan* NPs are inexpensive, biocompatible, antimicrobial, and biodegradable	Meng et al., 2019), Remya et al., 2022), Crini & Lichtfouse, 2019b)

activity which in turn enhances the reduction of the dyes (Khan et al., 2021b). Antioxidant activity is the free radical scavenging activity that corresponds to the antioxidant potential due to the negatively charged NP surfaces. These free radicals then attract the hydroxyl ions of the dye (Bhat et al., 2020b). TiO_2 possesses promising photocatalytic activity, it exists in three types of modifications, and studies illustrate that brookite and rutile perform the photocatalytic activity to a lesser extent than anatase (Dontsova et al., 2020).

Semiconductor oxide such as TiO_2 and SnO_2 possesses a broad band gap and utilizes only 5% solar light in the UV region. Therefore, there is a need to prepare PCs, which would be capable to utilize a major portion of solar light in the visible region (Pattnaik et al., 2018b). Researchers found a problem with the separation of NPs while using them as photocatalysts. The separation of NPs after catalysis proved an expensive, hard, and time-consuming task (Konstantinou & Albanis, 2004; Patil et al., 2019). To overcome these problems, researchers focused on preparing magnetic NPs that can be separated by using an external magnet and utilizing solar light effectively. The NPs made up of Au (Ayati et al., 2011; Liu et al., 2016; Oros-Ruiz et al., 2011), Ag (Mo et al., 2015; Namratha et al., 2013; Xiong et al., 2011), Fe_2O_3 (Jiang et al., 2013; Roushenas et al., 2016), Fe (Kozma et al., 2016; Zelmanov & Semiat, 2015), $Ni(OH)_2$ (El Hassani et al., 2019; Jayakumar et al., 2017; Nagajyothi et al., 2019; Yousefi et al., 2016), SiO_2 (Qin et al., 2018), SnO (Bhattacharjee et al., 2016; Elango et al., 2015, 2016; Kumar et al., 2018), TiO_2 (Barbosa et al., 2015; Raliya et al., 2017; Sahoo et al., 2012), ZnS (Yang et al., 2015), ZrO_2 (Bansal et al., 2015), CuO (Katwal et al., 2015; Mehr et al., 2018), $CoFe_2O_4$ (Kalam et al., 2018; Sundararajan et al., 2017), anatase $(TiO_2)_n$ (Gautam et al., 2016), and CdS (Darwish et al., 2016) are commonly used for water treatment. Among the salts of transition metals, Cr_2O_3 NPs show better results in the photocatalytic activity of dyes and pigments. $AgCrO_4$ NPs were prepared by using various methods including direct precipitation, electro-synthesis, and hydrothermal methods. In the visible region, these NPs show maximum results as PCs (Sarani et al., 2021). Fe_2O_3 NPs have been found the most excellent PCs due to their higher adsorption

capacity and ease in separation after catalysis (Giri et al., 2011; Vasantharaj et al., 2019). Moreover, nanomaterials have found applications in pollutant sensing, antibacterial activity, remediation of industrial effluents, and so on. The production of nanomaterials has been increased since 2011–2020 from 4000 to 58,000 tons, due to their diverse utilization in various fields (Sharma, 2009). CuO NPs show excellent results in photocatalytic degradation of CR dye by using cyanobacterium. It proved to be very less toxic and cheap method of dye degradation (Alsamhary et al., 2021). Azar et al. prepared CuO NPs having three different morphologies (nanosheets, nanorods, nanoleaves). This was associated with many hydroxyl ions, produced due to oxygen vacancies. These hydroxyl radicals act as scavengers and degrade the dye into inorganic smaller molecules (Sadollahkhani et al., 2014). Zhang et al. used $1gL^{-1}$ magnetic $Sr_5(PO_4)_3(OH)/Fe_3O_4$ NPs for 89% degradation of CR (Zhang et al., 2016). Titanate-supported intercalated CdS NPs showed complete color removal within 15 min (Sehati et al., 2016). Electron mediators like monometallic Ru NPs caused electrons to shift from the reducing agent to the characteristic bond (Azo bond) of CR; it caused an increase in degradation rate (Gupta et al., 2013). NPs which are a combination of two or three metals make an extraordinary catalyst for the breakdown of many azo compounds (Eskandarinezhad et al., 2021). Gd^{3+} doped cobalt ferrite NPs possessed greater removal ability (16 mg g^{-1}) than un-doped cobalt ferrite NPs (131 mg g^{-1}) (Zhao et al., 2014). Doping increases the absorption capabilities of cobalt ferrite NPs by generating unpaired electrons (Masunga et al., 2019). Similarly, ZnO shows better results in dye degradation when it is blended with polyvinyl pyrrolidone because the surface area and pore size of this composite are larger than the simple unmodified ZnO PC (Khan et al., 2020a). Silver NPs synthesized by using leaf extract of Chinese spinach (*Amaranthus gangeticus* Linn) were found to exhibit high catalytic potential (more than 50%) for decoloring CR (Kolya et al., 2015). The high pH value increased the production of OH^- ions which react to h^+ and form $OH^{\cdot}$ radicals. Thus, the higher number of $OH^{\cdot}$ radicals increased the photodegradation activity (Khairnar et al., 2018). At lower pH, an increased number of holes act as major oxidizing agents; as a result, photodegradation increased (Adam et al., 2018; Nezamzadeh-Ejhieh et al., 2015). Some dyes can be degraded by the use of UV irradiation only (Zhao et al., 2005). Kinetics studies illustrate that degradation of CR dye is followed by pseudo-1st-order reaction mechanism and CR dye degeneration shows maximum value of rate constant under the influence of UV light (Tama et al., 2020). Movahedi et al. showed that both PCs and UV irradiation are necessary for the effective degradation of CR. The pH of the solution containing dye and PCs greatly affects photocatalytic activity, because most semiconductor PCs show amphoteric behavior due to zero-point charge (ZPC) (6 for TiO_2) (Lizama et al., 2002; Zhao et al., 1998). Anionic dyes such as CR degraded effectively with PCs having low ZPC as at a high ZPC, the negative surface of the PCs repels sulfonated ions (Swarnalatha & Anjaneyulu, 2004). Besides, molecules of the dyes tend to aggregate in acidic or highly acidic media and form tautomer (Fig. 9) due to which photocatalytic activity was not completed (Movahedi et al., 2009b; Vidya et al., 2017b). The optimum concentration of catalyst increased the photocatalytic activity. The higher the number of catalysts caused the solution to become turbid, which results in light scattering phenomena (Adam et al., 2018; Ajoudanian & Nezamzadeh-Ejhieh, 2015; Ramaswamy et al., 2008; Ullah et al., 2018). As the concentration of dye increased by keeping the catalyst amount constant, photodegradation activity decreased due to saturation of active sites by the given catalyst (Khairnar et al., 2018). At a higher concentration of dye, the contact time increased and photocatalytic activity decreased due to light hindering property of dye molecules (Bansal & Sud, 2011). For maximum degradation, there is an optimum duration of time above; this time degradation rate remains constant due to saturation of active sites of the catalyst by dye molecules (Bansal & Sud, 2011; Khairnar et al., 2018; Ullah et al., 2018). One of the important green chemistry technologies is the photodegradation of organic dyes. PCs are required for the degradation of dye. Photodegradation by NPs bears many advantages; the most important is the complete degradation of dyes (Banerjee et al., 2019). Due to the high surface area, active sites, small size, nontoxicity, high chemical stability

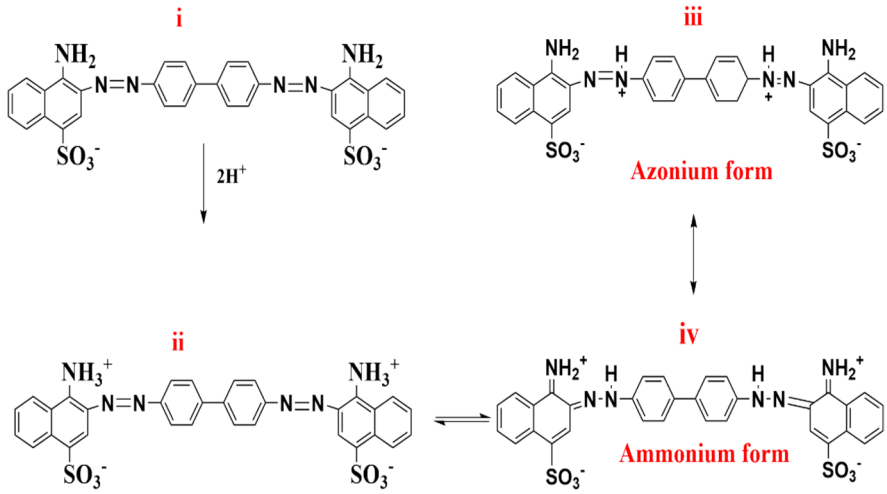


Fig. 9 Protonation of CR in the acidic medium (Movahedi et al., 2009b)

(Gunjakar et al., 2011), low cost, and high catalytic potential, nanomaterials have proved themselves as efficient adsorbents (Raval et al., 2016).

The catalytic activity of bismuth ferrite (BFO) NPs was found to increase as preparation temperature increased from 350 to 500 °C and then decreased as temperature increased (Pattnaik et al., 2018b). The green synthesis of NPs using plants and microorganisms is the other choice for researchers as it proves less hazardous. However, biosynthesis is a very slow process, which is considered the major limitation of this method (Gadd et al., 1989; Saifuddin et al., 2009). Plant extract acts as a green reducing agent that helps to form metal nanoparticles (MNPs) via the conversion of metal ions into atoms and finally into MNPs by coagulation (Maryami et al., 2016). S.A Moon et al. prepared NPs of CuO, Ag, and MnO_2 by using the extract of plant *Kalopanax pictus*; photocatalytic potential for CR of these NPs showed that MnO_2 is more effective in the removal of CR from the aqueous media than that of CuO and Ag NPs (Moon et al., 2018).

7.1 Mechanism of Photocatalysis

Semiconductors are mostly used as PCs. They are considered highly stable and environment-friendly (Rajendran et al., 2021b). Efficiencies of various photocatalysts have been mentioned in Table 6. The process of catalysis is carried out with the help of charge carriers as described in Fig. 10. The number of charge carriers can be increased with the help of doping (Terna et al., 2021b). The photo-generated hole/electron has possibilities of recombining again or may combine with the adsorbate (dye). Photo-generated electron in the CB and h^+ in the VB served as potential oxidizing and reducing agents respectively. As a result of oxidation–reduction reactions, adsorbate (dye) is converted into small nontoxic inorganic substances (Bhat et al., 2020b; Pattnaik et al., 2018b; Vidya et al., 2017b). The separation of the charge carrier is an important factor that contributes to the efficiency of PCs. This separation can be achieved by different methods including metallization and doping (Arkhipov et al., 2005b). Hydroxyl radical $OH^{\cdot}$ is the main active agent for the photodegradation of organic substances, although the process mechanism is different in the case of different dyes and NPs. In the photodegradation process as described in Fig. 11, aromatic rings were attacked by free $OH^{\cdot}$ radicals, resulting in hydroxylated intermediates and finally release of CO_2 by a series of photo-Kolbe reactions. Gaseous nitrogen is released in the photocatalytic degradation of azo dyes, which is an ideal condition for the mineralization of nitrogen-bearing pollutants (Movahedi et al., 2009b). It is reported that heteroatoms such as sulfur are converted into nontoxic sulfate SO_4^{-2} ions. It is reported that heteroatoms such as sulfur are converted into nontoxic sulfate SO_4^{-2} ions (Lachheb et al., 2002).

Table 6 Comparison of efficiencies of reported materials for photodegradation of CR

Catalyst	Preparation method	Amount of catalyst	Dye (mgL^{-1})	Irradiation source	Time (min)	% degradation	References
NiO NPs	Co-precipitation method	20 mg	30	UV light	20	72	Bhat et al. (2020b)
CuO nanosheets	Co-precipitation	0.05 g/100 mL	20	UV light	210	12	Sadollahkhani et al. (2014)
CuO nanoleaves	Co-precipitation	0.05 g/100 mL	20	UV light	210	48	Sadollahkhani et al. (2014)
CuO nanorods	Co-precipitation	0.05 g/100 mL	20	UV light	210	67	Sadollahkhani et al. (2014)
$Sr_5(PO_4)_3(OH)/Fe_3O_4$	Solvothermal	$1gL^{-1}$	500	UV light	60	89	Zhang et al. (2016)
CdS/Ti_4O_9 NPs	Ultrasonication	0.05 g	50	Sunlight	60	-	Sehati et al. (2016)
ZnO powder	-	$0.5gL^{-1}$	5.7	UV light	8	-	Movahedi et al. (2009b)
AC-AV NPs	-	$1gL^{-1}$	100	No light source	20	-	Khaniabadi et al. (2017)
Bismuth ferrite (BFO) NPs	Solid-state reaction	$1gL^{-1}$	10	Sunlight	60	77	Pattnaik et al. (2018b)
ZnO NPs	Green synthesis	$0.24gL^{-1}$	20	UV light	60	>90	Vidya et al. (2017b)
Nb_2O_5	Hydrothermal	$1gL^{-1}$	10	UV light	120	90	Khairnar et al. (2018)

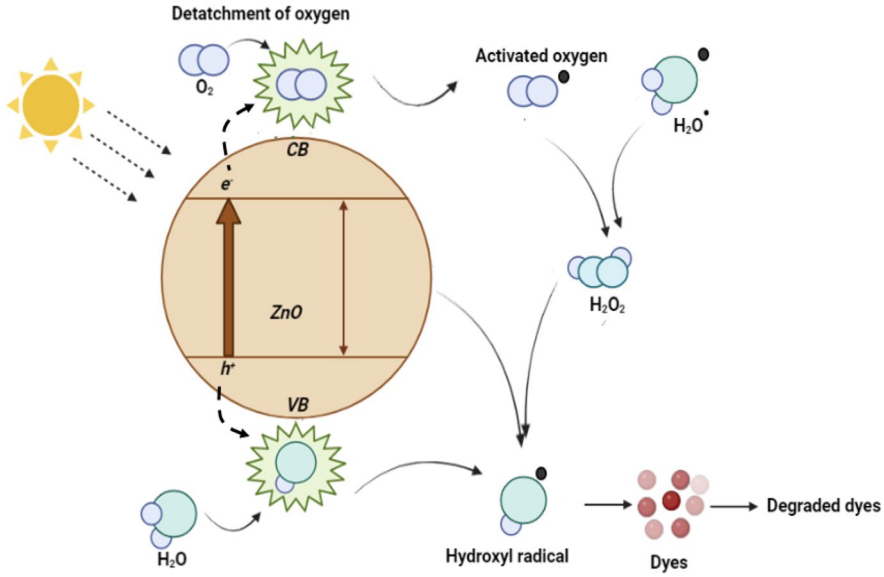


Fig. 10 Mechanism of photodegradation of CR (Hairom et al., 2015)

8 Conclusion

Synthetic dyes have replaced natural pigments due to their extensive photostable and thermostable properties. Various conventional methods for removing dyes from wastewater, such as coagulation, ozonation, ultra-filtration, and AOP, are now prohibited due to their high cost, high energy

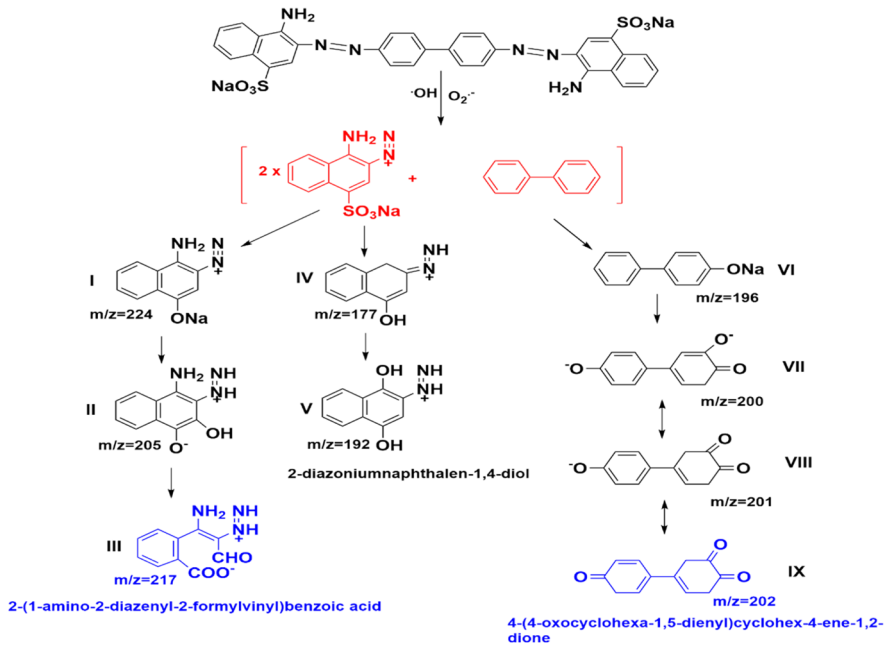


Fig. 11 The proposed mechanism of photodegradation of CR

consumption, and high sludge production. Different useful methods for the conversion of recalcitrant CR dye into less toxic degraded products have been explicated. CR dye is an azo dye that is a complex aromatic structure attached with –N = N– bonding. Because of its large aromatic structure, it is resistant to natural degradation and has a harmful effect on the ecosystem by reducing the photosynthetic activities of aquatic flora, causing eutrophication. Auxochromes and chromophores attached to dyes are carcinogenic when they react with heavy metals. The preceding discussion aims to collect data on the degradation of CR dye using fungi and bacterial species, as well as the photodegradation method using NPs. The presence of various lysosomal enzymes in several bacterial species has helped in the discovery of dye degradation in a less toxic way. The extraction of bacterial cultures is easily done from the soil, water, or contaminated food. The dye pretreated with sonication by using ultrasounds reduced the degradation time by *Bacillus-Aspergillus* than the non-pretreated dye sample. Anaerobic, anoxic, and aerobic are different conditions required by bacterial species to degrade dyes. Fungal degradation of dyes is becoming a crucial method due to the presence of extracellular enzyme activity in fungi. Among fungi, white-rot fungi and many marine fungal species are proved to be very efficient in the degradation process of dyes. Experiments have been done to check the effect of nutrient sources in fungal culture. Inorganic media are known as the best source of carbon for fungal growth resulting in the efficient decoloration of CR dye. *Aspergillus* sp. showed efficient degradation of dye at optimum shaking speed. At high shaking speed, the enzymatic activity of manganese peroxidase (MnP) and lignin peroxidase (LiP) in fungi is maximum which results in efficient degradation of CR dye. In like manner, *Aspergillus* sp. showed maximum extracellular activity of MnP and LiP at optimum temperature range. One of the best and most prominent methods is photocatalytic degradation of dyes by using NPs. It is becoming a vital method because of its capability to degrade the dye completely, high stability, nontoxic nature, low cost, high surface area, active sites, and high catalytic potential. Iron oxide NPs are found to be the most efficient PCs due to their high maximum adsorption capacity. Experiments have shown that CuO nanorods are more efficient in CR photodegradation due to the presence of hydroxyl ions, which act as scavengers and break the dye into small inorganic ions. Similarly, silver nanoparticles have a high catalytic potential for degrading pigment. PCs and ultraviolet (UV) irradiation have both been shown in studies to be very effective in CR degradation.

Data Availability The data that support the findings of this study are available on request from the corresponding author.

Declarations

Conflict of Interest The authors declare no competing interests.

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