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Bioengineering for Decolorization of Synthetic Dyes in Textile Effluents using Microbial Enzymes

Aparajita Sen[1] and Arti Nigam[*2]

[1] Department of Genetics, University of Delhi, South Campus, New Delhi, India, senaparajita99@gmail.com
[*2] Department of Microbiology, Institute of Home Economics, University of Delhi, New Delhi, India, arti.nigam@ihe.du.ac.in

Abstract: **With the increasing use of synthetic dyes in textile industries, there has been a simultaneous increase in the levels of water pollution, as a result of release of effluents from these textile industries. The effluents constitute majorly of synthetic dyes which are toxic and harm aquatic organisms. Thus, we need to treat the effluents prior to release into the environment. The available physio-chemical methods have limitations when applied at large scale, and thus biological methods are the most suitable. Biological agents may be biomass or purified enzymes. Dye decolorization is achieved mostly by either/both biodegradation and biosorption. In biodegradation, the dye is converted to a less toxic product by microbial enzymes, and in biosorption the dye particles are adsorbed on the surface of the microbial cells. In this review, there is a compilation of the efficiency of dye decolorization by enzymatic degradation of various azo and anthraquinone dyes by microbial enzymes.**

Index Terms: **Decolorization of Synthetic Dyes, Dye Bioremediation, Textile Effluents, Microbial Enzymes, Peroxidases**

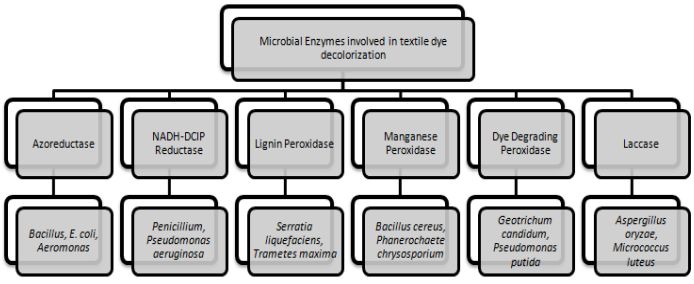

Fig. 1. Microbial Enzymes involved in Dye Decolorization

I. Introduction

A dye is a synthetic colored chemical substance, which when applied to fibers, colors them permanently. This color does not fade even upon microbial attack, exposure to light, salts, sweat, water and most of the chemicals (Rai et al, 2005). Due to the expansion of the global textile industry, a peak in usage of synthetic dyes has been observed (Sen et al, 2021a), and this has also resulted in increased pollution because of wastewater containing dye contaminants (Pandey et al, 2007). Treating these volumes of wastewater is needed; else the water bodies into which they are released will be rendered unusable due to presence of dyes. Based on processing stages, there are different wastewater characteristics, like dissolved oxygen (DO), pH, inorganic and organic chemical contents, etc (Saratale et al, 2009). Careless release of these effluents into the aqueous ecosystems harms the aquatic life. Moreover, many synthetic dyes are toxic, carcinogenic, and mutagenic (Dawkar et al, 2010).

There are many physical and chemical methods for the removal of synthetic dyes from effluents of textile industries (Gupta, 2009). These techniques produce large amounts of sludge which too requires further safe disposal, thus slowing down efforts to eradicate water pollution, particularly in developing countries (Khandare et al, 2013). Thus, economical and ecofriendly alternatives are required. The biological alternatives have many advantageous characteristics. (Jadhav et al, 2009). This technique of using living organisms like bacteria, algae, fungi, actinomycetes, yeasts or dead biomass for the biodegradation/biosorption of textile dyes and other xenobiotics is called BIOREMEDIATION.

Removal of color by either conversion of a chromophore to a non-chromophore is termed as “decolorization”. While biodegradation is the breakdown of the dye molecule by enzymes, adsorption of the dye on the surface of the support also causes decolorization. The latter does not cause a change in the composition of a dye. Decolorization by degradation, however,

* Corresponding Author

always results into the formation of a new product (Gao et al, 2010). A number of advanced and developed instrumental techniques are available for verification of Decolorization by degradation including Atmospheric pressure chemical ionization (APCI), Gas Chromatography/Mass Spectrometry (GC/MS), High-Performance Liquid Chromatography (HPLC), and Fourier Transform Infra-Red spectroscopy (FTIR) (Jadhav et al, 2010). These techniques have revolutionized the chemical analysis procedures and give accurate information about the structure, molecular orientation and functions of the dye molecule, which plays a key role in the verification and predictions of steps that are associated with decolorization (Gao et al, 2010).

II. MICROBIAL DYE DECOLORIZATION

Most microbes are able to produce large amounts of enzymes like peroxidases, laccases, azoreductases, etc, as shown in Fig. 1. These enzymes play a role in degrading most azo and anthraquinone dyes to colorless and less toxic compounds by various mechanisms, as has been discussed in Section 2. Many white rot fungi, like *Phanerochaetae chrysosporium* and *Trametes versicolor* can degrade synthetic dyes (Sharma et al, 2009; Bibi & Bhatti, 2012). Some bacteria that possess this capability are *Serratia marcescens* (Gusmanizar et al, 2016), *Serratia liquefaciens* (Haq & Raj, 2018), *Pseudomonas aeruginosa* (Phugare et al, 2011), *Acinetobacter radioresistens* (Ramya et al, 2010), *Brevibacterium* (Franciscon et al, 2012), *Aeromonas hydrophila* (Thanavel et al, 2019), *Bacillus subtilis* (Barathi et al, 2020), *Bacillus cereus* (Sheela & Sadasivam, 2020), *Anoxybacillus* (Wang et al, 2020) etc. Sporulating fungi like *Aspergillus* (Asses et al, 2018), *Geotrichum* sp. (Rajhans et al, 2020) and *Penicillium* (Ayla et al, 2018) have also been reported for the decolorization of synthetic dyes. Actinomycetes like *Streptomyces chromofuscus* can also decolorize synthetic dyes (Dong et al, 2019).

There are many parameters that affect the efficiency of dye decolorization, such as properties of the microbes, whether the inoculum is of a pure culture, mixed culture, or a consortium, whether the microbes are aerobic, anaerobic, or both. Properties of the dye, such as structure, concentration and toxicity also determine decolorization efficiency. Apart from these, efficiency of dye decolorization is also influenced by physiological conditions, such as temperature, pH, inoculum concentration, dissolved oxygen, incubation time etc (Garg & Tripathi, 2017).

III. ADVANTAGES OF MICROBIAL METHODS OF DYE DECOLORIZATION OVER OTHER METHODS

Advantages of bioremediation include conversion of the organic compounds to non-toxic products (carbon dioxide and water), sustainability, low cost and the ease of operation (Al-Tohamy et al, 2020). These techniques are also environment friendly, and do not produce sludge, unlike as in the case of the physiochemical methods (Khandare et al, 2013). Microbes are also easier and faster to grow, and are less likely to cause much technical difficulties during the decolorization process (Karim et al, 2018).

Physiochemical methods have shortcomings such as high operating cost, large quantities of sludge and interferences from the other constituents of the wastewater. Biological methods, however, are economic and have stable effects. Thus, biological methods have been widely used, and more specifically, microbial methods are the most suitable (Holkar et al, 2016; Nouren et al, 2017).

The available physicochemical methods, such as flocculation/coagulation, adsorption, precipitation, oxidation, membrane extraction, electrolysis and advanced oxidation processes (Imran et al, 2015). These methods are effective, but also very chemical/energy intensive. Moreover, chemical methods introduce further chemicals which may be toxic themselves. Most physical methods concentrate pollutants, which need further safe disposal or treatment, further increasing the cost of treatment. Microbes, on the other hand, can completely degrade the dyes instead of concentrating, and thus reduce the time, labor and cost required (Sandhya et al, 2007). Due to these drawbacks, microbial methods are considered more specific, effective and safe, as they completely convert the organic pollutants to non toxic stable end products. Some microbes also can adsorb the pollutants, and thus are the greener alternatives (Wariishi et al, 2002).

IV. ENZYMATIC DEGRADATION OF SYNTHETIC TEXTILE DYES

Dyes can be decolorized by either reductive enzymes such as azoreductases, or oxidative enzymes like peroxidases and laccase (Singh et al, 2015; Sheela & Sadasivam, 2020).

A. Azoreductases

Azoreductases are flavoproteins that use electron donors (NADH/ NADPH/ FADH) to reduce the azo bond of azo dyes (Russ et al, 2000) and convert them into their corresponding colorless aromatic amines. The breakdown of the azo bond occurs at the bacterial cell membrane, either intracellularly or extracellularly. Thus these enzymes are potent decolorization agents of textile effluents (Ramya et al, 2010; Dong et al, 2019; Sheela & Sadasivam, 2020).

Azoreductases are produced by a wide variety of microorganisms, such as *Pigmentiphaga kullae* K24 (Blumel & Stolz, 2003), *Xenophilus azovorans* KF46F (Blumel et al, 2002), *Enterococcus faecalis* (Chen et al, 2004), *Staphylococcus aureus* (Chen et al, 2005), *Bacillus* sp. OY1-2 (Suzuki et al, 2001), and *Rhodobacter sphaeroides* (Bin et al, 2004). It was reported that a recombinant strain of *Escherichia coli* expressing the *X. azovorans* KF46F azo B gene showed around 50X higher azoreductase activity than by *X. azovorans* KF46F **(**Blumel et al, 2002). Azoreductase is expressed by many microbes that are a

part of the human gut microbiota, such as *Clostridium*, *Pseudomonas*, *Bacillus*, *Geobacillus*, *Lysinibacillus*, *Enterococcus* and *Eubacterium* (Zahran et al, 2019).

There have been lots of applications of azoreductases in textile dye decolorization which have been reported by various researchers. In all the cases, various azo and anthraquinone dyes were decolorized up to 64-100%, which is significant for the treatment of textile effluents. Some such data has been summarized in Table I.

Table I. Decolorization of various azo dyes by microbial Azoreductases

Producer Microbe	Dye Decolorized	% Decolorized*	Reference
Acinetobacter radioresistens	Acid Red	>70	Ramya et al (2010)
Aeromonas hydrophila SK 16	Acid Fast Yellow MR	91.25	Thanavel et al (2019)
Alcaligenes sp. AA09	Reactive Red BL	100	Pandey & Dubey (2012)
Bacillus cereus SKB12	Reactive Black 5	88.7	Sheela & Sadasivam (2020)
Bacillus lentus BI377	Reactive Red 141	99.11	Oturkar et al (2013)
Bacillus megaterium	Red 2G	64.89	Khan (2011)
Bacillus strain SF	Reactive Black 5	86	Maier et al (2004)
	Mordant Black 9	38	
Bacillus subtilis ORB7106	Methyl Red	98	Leelakriangsak & Borisut (2012)
Brevibacterium sp. strain VN-15	RY107	98	Franciscon et al (2012)
Consortium of *Providencia* sp. SDS and *Pseudomonas aeuroginosa* BCH	Red HE3B	100	Phugare et al (2011)
Enterococcus faecalis	Methyl Red	100	Chen & Ting (2015)
Enterococcus gallinarum	Direct Black 38	100	Bafana et al (2009)
Escherichia coli JM109 (pGEX-AZR)	Direct Blue 71	100	Jin et al (2009)
Mutant *Bacillus* sp. ACT2	Congo Red	30	Gopinath et al (2009)
Proteus sp.	Congo Red	67	Perumal et al (2012)
Pseudomonas aeruginosa	Remazol Orange	94	Sarayu & Sandhya (2010)
Staphylococcus aureus	Methyl Red	100	Chen et al (2005)
Streptomyces sp. S27	Methyl Red	99	Dong et al (2019)
Xenophilus azovorans KF46F	Acid Orange 7	100	Blumel et al (2002)

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B. NADH-DCIP Reductase

NADH-DCIP Reductase is a part of the multifunctional oxidase system of bacteria, and is involved in the bioremediation of xenobiotics (Salokhe & Govindwar, 1999). Using NADH, it reduces DCIP (2,6-dichloroindophenol), turning its blue color to colorless. The nonspecific reductase which was induced significantly during biodegradation of Malachite Green was called MG Reductase. It uses NADH and reduces Malachite Green to Leucomalachite Green (Parshetti et al, 2006).

This enzyme is produced by many bacteria like *Pseudomonas aeuroginosa* BCH (Jadhav et al, 2009), *Bacillus subtilis* (Barathi et al, 2020) etc., and fungi like *Penicillium* sp. YW 01 (Yang et al, 2011), *Sterigmatomyces halophilus* SSA1575 (Al-Tohamy et al, 2020), *Achaetomium strumarium* (Bankole et al, 2018), *Perenniporia subacida* (Si et al, 2014) etc. Many consortia have proven to be better in decolorizing textile dyes than the individual microbial strains. For example, a consortium consisting of *Micrococcus glutamicus* NCIM-2168 and *Proteus vulgaris* NCIM-2027 could decolorize Green HE_4BD **by** 86% (Saratale et al, 2010). A consortium of *Pseudomonas aeuroginosa* BCH and *Providencia* sp. SDS decolorized Red HE_3B completely (Phugare et al, 2011). A consortium of *Providencia rettgeri* strain HSL1 and *Pseudomonas* sp. SUK1 decolorized Direct Red 81 by 99% (Lade et al, 2015).

Many researchers have reported the role of NADH-DCIP reductases in textile dye decolorization. Percentage decolorization ranged from 86-100%, demonstrating the possibility of use of this enzyme more widely for effluent treatment. Some such examples have been compiled in Table II.

Table II. Decolorization of dyes by microbial NADH-DCIP Reductase

Producer Microbe	Dye Decolorized	% Decolorized*	Reference
Achaetomium strumarium	Acid red 88	99	Bankole et al (2018)
Bacillus circulans BWL1 061	Methyl Orange	99.22	Liu et al (2017)
Bacillus subtilis	Reactive Blue 160	100	Barathi et al (2020)
Proteus vulgaris NCIM-2027 and *Micrococcus glutamicus* NCIM-2168	Green HE_4BD	86	Saratale et al (2010)
Providencia rettgeri strain HSL1 and *Pseudomonas* sp. SUK1	Direct Red 81	98-99	Lade et al (2015)
Consortium of *Providencia* sp.	Red HE_3B	100	Phugare et al (2010)

SDS and *Pseudomonas aeuroginosa* BCH			
Penicillium sp. YW 01	Malachite Green	98.23	Yang et al (2011)
Perenniporia subacida	Neutral Red	96.56	Si et al (2014)
Pseudomonas aeuroginosa BCH	Orange 39	93.06	Jadhav et al (2010)
Pseudomonas monteilii ANK	Scarlet RR	97	Kabra et al (2013)
Sterigmatomyces halophilus SSA1 575	Reactive Black 5 (RB5)	100	Al-Tohamy et al (2020)

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C. Lignin Peroxidase (LiP)

LiP is an oxidoreductase, and specifically acts on peroxide as an electron acceptor, and thus is called as peroxidase. It is a part of the broad category of ligninases. LiP catalyzes multiple oxidations in the lignin (or lignin-related compounds) side chains by removal of one electron at a time, to form reactive radicals (A•+), where A is the substrate, or dye to be decolorized (Tien & Kirk, 1983; Kersten et al, 1990).

LiP is produced by microbes like *Providencia* sp. SRS82 (Agrawal et al, 2014), *Serratia liquefaciens* (Haq & Raj, 2018), *Bacillus cereus* SKB12, *Phanerochate chrysosporium* and other white-rot fungi such as *Trametes maxima*, *Phanerochaete sordida*, *Phlebia radiata* and *Phlebia tremellosa* (Harazono et al, 2003). Some other fungi that use LiP for decolorizing dyes are *Aspergillus niger* (Asses et al, 2018), *Ganoderma lucidum* (Shaheen et al, 2017), *Stereum ostrea* (Usha et al, 2014) etc.

There are many reported uses of bacterial as well as fungal LiP in dye decolorization, showing 61-100% decolorization of many azo dyes. Some of such reports have been summarized in Table III.

Table III. Decolorization of dyes by microbial Lignin Peroxidase

Producer Microbe	Dye Decolorized	% Decolorized*	Reference
Acinetobacter calcoaceticus NCIM 2890	Methyl Red	98	Godhake et al (2009)
Aspergillus niger	Congo Red	>97	Asses et al (2018)
Bacillus cereus SKB12	Reactive Black 5	88.7	Sheela & Sadasivam (2020)
Ganoderma lucidum IBL-05	Red C4BLN	93	Shaheen et al (2017)
Kocuria rosea MTCC 1532	Methyl Orange	100	Parshetti et al (2006)
Phanerochaete chrysosporum	Ranocid Fast Blue	83	Verma & Madamwar (2002)
Phanerochate chrysosporium	Rhodamine B	91	Lan et al (2006)
F. F. Lombard ME446			
Phanerochaete sordida	Reactive Red 120	90.6	Harazono et al (2003)
Phlebia tremellosa	Remazol Red	100	Kirby et al (2000)
Polyporus ostreiformis	Congo Red	99	Dey et al (1994)
Providencia sp. SRS82	Acid Black 210	90	Agrawal et al (2014)
Serratia liquefaciens	Azure B	90	Haq & Raj (2018)
Stereum ostrea	Crystal Violet	90	Usha et al (2014)
Trametes maxima LE130	Reactive Black 5	61	Levin et al (2019)

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D. Manganese Peroxidase (MnP)

MnP is another peroxidase that can be broadly classified as a ligninase. The glycoprotein consists of heme, and oxidizes Mn^{2+} to Mn^{3+}, which is an electron donor. Mn^{3+}, in turn, can oxidize many phenolic substrates. The reaction it catalyzes has A as the substrate, or dye to be decolorized, and A•+ is the oxidized reactive radical (Eibes et al, 2006).

MnP is produced by many white-rot fungi like *Phanerochaete chrysosporium*, *Phanerochaete sordida*, *Trametes polyzona* etc (Sharma et al, 2009; Lueangjaroenkit et al, 2019). There are eleven different isoforms of MnP in *Ceriporiopsis subvermispora* (Chmelová & Ondrejovič, 2016). Bacteria like *Bacillus cereus* SKB12 also produce MnP (Sheela & Sadasivam, 2020).

There have been many reports of the role of MnP in decolorization of textile dyes, showing 63-100% decolorization of several azo dyes. Some such reports have been summarized in Table IV.

Table IV. Decolorization of dyes by microbial Manganese Peroxidase

Producer Microbe	Dye Decolorized	% Decolorized*	Reference
Bacillus cereus SKB12	Reactive Black 5 dye	88.7	Sheela & Sadasivam (2020)
Bjerkandera adusta	Reactive Violet 5	87	Heinfling et al (1998)
Bjerkandera sp. strain BOS55	Orange II	>85	López et al (2004)
Ceriporiopsis subvermispora ATC C 90467	Malachite Green	87.8	Chmelová & Ondrejovič (2016)
Daedaleopsis confragosa	Green HE4BD	90.08	Manawadi et al (2019)
Ganoderma lucidum IBL-05	Sandal-fix Black CKF	95.7	Bilal & Asgher (2015)
Irpex lacteus	Remazol Brilliant Blue R (RBBR)	100	Svobodová et al (2006)

Irpex lacteus F17	Direct Sky Blue 5B	63.75	Duan et al (2018)
Phanerochaete chrysosporium	Bromophenol blue	99.3	Svobodová et al (2006)
Phanerochaete chrysosporium	Orange II	85	Sharma et al (2009)
Phanerochaete sordida	Reactive Red 120	90.6	Harazono et al (2003)
Schizophyllum sp. F17	Orange IV	76	Yao et al (2013)
Strain L-25 (white rot fungus)	Reactive Orange 16	99.6	Kariminiaae-Hamedaani et al (2007)
Trametes polyzona KU-RNW027	Remazol Brilliant Blue	100	Lueangjaroenkit et al (2019)

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E. Dye Degrading Peroxidase (DyP)

The DyP is a new class of heme peroxidases which show no homology in structure or sequence to other microbial peroxidases. They have wide substrate specificity. They can function optimally in low pH conditions, and oxidize the typical substrates of peroxidases, in addition to other synthetic high redox potential anthraquinone dyes, which cannot be converted by the other peroxidases (Min et al, 2015).

DyP is produced by fungi like *Auricularia auricula-judae* (Liers et al, 2013), *Geotrichum candidum* (Rajhans et al, 2020), *Irpex lacteus* (Duan et al, 2018), *Pleurotus ostreatus* (Cuamatzi-Flores et al, 2019) etc., and bacteria like *Pseudomonas putida* and *Bacillus subtilis* (Santos et al, 2014) etc. Some studies reporting the role of DyP in dye decolorization have been summarized in Table V.

Table V. Decolorization of dyes by microbial Dye Peroxidase

Producer Microbe	Dye Decolorized	% Decolorized*	Reference
Auricularia auricula-judae	Azure B	100	Liers et al (2013)
Geotrichum candidum	Methyl Orange	94.6	Rajhans et al (2020)
Irpex lacteus F17	Reactive violet 5	92.16	Duan et al (2018)
Pleurotus ostreatus	Acid Blue 129	77	Cuamatzi-Flores et al (2019)
Pseudomonas putida	Direct Red 5B	81	Khandare et al (2013)

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F. Laccases

Laccases or multicopper oxidases (MCO**)** are Polyphenol Oxidases (Birhanli & Yesilada, 2006; Giardina et al, 2010; Arora & Sharma, 2010), which can degrade and decolorize phenolic compounds as well as aromatic azo dyes. They can oxidize aromatic amines using Copper (II) as the mediator (Sudha et al, 2014; Singh et al, 2015; Mehta et al, 2016).

Laccase is mainly produced by wood-degrading fungi such as *Tinea versicolor* (Mostafa et al, 2019), *Trametes hirsuta* (Yanto et al, 2019), *Cerrena unicolor* (Michniewicz et al, 2008), *Ceriporiopsis subvermispora* (Chmelová & Ondrejovič, 2016) etc. Bacterial laccase was first reported in a bacterium called *Azospirillum lipoferum* (Singh et al, 2007). Since then, laccase activity has also been demonstrated in other bacteria such as *Bacillus subtilis*, *Streptomyces griseus*, and *Thermus thermophilus* etc (Kumari et al, 2018).

Laccases are the most efficient and most applied enzymes in bioremediation of textile dyes. There are many applications of laccases in the decolorization of dyes, resulting in 75-100% decolorization in most cases. Some of such studies have been compiled in Table VI.

Table VI. Decolorization of dyes by microbial Laccases

Producer Microbe	Dye Decolorized	% Decolorized*	Reference
Aeromonas sp. DH-6	Methyl Orange	100	Du et al (2015)
Anoxybacillus ayderensis SK3-4	Direct green 6	100	Wang et al (2020)
Aspergillus oryzae	Drimaren Blue	80-90	Teixeira et al (2010)
Armillaria sp. F022	Reactive Black 5	80	Hadibarata et al (2012)
Bacillus cereus SKB12	Reactive Black 5	88.7	Sheela & Sadasivam (2020)
Ceriporiopsis subvermispora ATCC 90467	Malachite Green	87.8	Chmelová & Ondrejovič (2016)
Cerrena unicolor	Acid Red 27	100	Michniewicz et al (2008)
Coprinopsis cineria	Methyl Orange	47.6	Tian et al (2013)
Coprinus plicatilis	Turquoise Blue HFG	100	Akdogan et al (2014)
Coriolopsis sp. (1c3)	Crystal Violet, Methyl Violet, Cotton Blue, Malachite Green	94, 97, 91 and 52 respectively	Chen & Ting (2015)
Curvularia sp.	Congo Red	100	Senthilkumar et al (2015)
Dichomitus squalens	Orange G, Remazol Brilliant Blue R (RBBR)	100 and 92 respectively	Eichlerová et al (2007)
Funalia trogii ATCC 200800	Crystal Violet	38	Yesilada et al (1995)
Ganoderma lucidum E47 strain	Bromocresol purple	56	Palazzolo et al (2019)
Ganoderma sp.	Methyl Orange	>90	Sun et al (2012)
Ganoderma sp. En3	RBBR, Indigo Carmine, Methyl	66-82, >93.4, >83 respectively	Lu et al (2016)

	Green		
Ganoderma sp. En3	Reactive Orange 16	95.1	Ma et al (2014)
Ganoderma weberianum TZC1	Indigo dye	92	Tian et al (2013)
Geobacillus catenulatus MS5	Congo Red	99	Verma & Shirkot (2014)
Geobacillus stearothermophilus	Indigo carmine, Congo Red, Brilliant Green	99, 98, 60 respectively	Mehta et al (2016)
Immobilized *Trametes pubescens, Pleurotus ostreatus*	Remazol Brilliant Blue R, Reactive Blue 49	>95	Chen & Ting (2015)
Irpex lacteus	Black Dycem	90	Baccar et al (2011)
Lentinus polychrous	Congo Red	75	Suwannawong et al (2010)
Micrococcus luteus	CI Acid Black 210	96.4	Kanagaraj et al (2015)
Oudemansiella canarii	Congo Red	80	Iark et al (2019)
Penicillium sp.	Vat brown - 5	75	Ayla et al (2018)
Peniophora cinerea	Textile Industry Effluent	54.6	Moreira et al (2014)
Pichia pastoris	Crystal Violet	90.7	Wang et al (2018)
Pleurotus ostreatus	Synazol Red HF6BN	96	Ilyas et al (2012)
Pleurotus ostreatus	Remazol Brilliant Blue R	80	Palmieri et al (2005)
Pleurotus ostreatus MTCC 142	Crystal Violet	92	Kunjadia et al (2012)
Pleurotus ostreatus URM 4809	Remazol brilliant blue R	86	Simões et al (2019)
Pleurotus ostreatus, P. sapidus, P. florida	Coralene Golden Yellow, Coralene Navy Blue, Coralene Dark Red	88, 92, 98 respectively for all dyes	Kunjadia et al (2016)
Podoscypha elegans	Rose Bengal	70.41	Pramanik & Chaudhari (2018)
Providencia rettgeri strain HSL1	C.I. Reactive Blue 172 (RB 172)	98-99	Lade et al (2015)
Providencia sp. SRS82	Acid Black 210 triazodye	90	Agrawal et al (2014)
Pseudomonas desmolyticum NCIM 2112	Direct Blue 6, Green HE4B and Red HE7B	100 for all three	Kalme et al (2009)
Pycnoporus sanguineus	Crystal Violet	49.4	Sulaiman et al (2013)
Pycnoporus sanguineus	Trypan Blue	70	Annuar et al (2009)
Serratia liquefaciens	Azure B	>90	Haq & Raj (2018)
Shewanella oneidensis (MFC)	Acid Orange 7	80.4	Mani et al (2019)
Thelephora sp.	Orange G	19	Selvam et al (2003)
Trametes hirsuta EDN084	Direct Blue	85	Yanto et al (2019)
Trametes versicolor	Reactive Black 5	42.78	Bibi & Bhatti (2012)
Trametes versicolor strain 1	Reactive Blue 4	90	Yemendzhiev et al (2009)
Trametes trogii	Textile Factory effluent	81	Khlifi et al (2010)

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G. Other Enzymes

There are some enzymes which are not very common in decolorization of textile dyes. These include tyrosinases, which convert monophenols to o-diphenols, and further to o-quinones. Some microbes producing these enzymes, and aiding in bioremediation of textile effluents are *Bacillus cereus* SKB12 (Sheela & Sadasivam, 2020), *Providencia sp.* SRS82 (Agrawal et al, 2014), *Kurthia huakuii* LAM0618 (Guo et al, 2016) etc. Veratryl Alcohol Oxidases are another class of such enzymes that oxidize veratryl alcohol to veratraldehyde, reducing O_2 to H_2O_2, which is used by peroxidases to further degrade the dyes. *Providencia rettgeri* strain HSL1, *Pseudomonas* sp. SUK1 and *Comamonas* sp. UVS have been reported to decolorize dyes via production of Veratryl Alcohol Oxidases (Jadhav et al, 2009; Lade et al, 2015). Superoxide Dismutases and Catalases also play a minor role in dye decolorization, and have been reported in *Lysinibacillus* sp. by certain researchers (Bedekar et al, 2014).

These enzymes have been reported to decolorize various azo dyes by 88-100% in most cases. Some of such reports have been summarized in Table VII.

Table VII. Decolorization of dyes by other microbial enzymes

Enzyme	**Producer Microbe**	**Mode of Action**	**Dye Decolorized**	**% Decolorized***	**Reference**
Polyphenol Oxidase/ Tyrosinase	*Bacillus cereus* SKB12	Catalyses o-hydroxylation of monophenols to o-diphenol and oxidation of o-diphenols to o-quinones	Reactive Black 5	88.7	Sheela & Sadasivam (2020)
	Providencia sp. SRS82		Acid Black 210 triazodye	90	Agrawal et al (2014)
	Kurthia huakuii LAM0618		Ethyl Violet	94	Guo et al (2016)
Veratryl Alcohol Oxidase	*Providencia rettgeri* strain HSL1	Oxidizes veratryl alcohol to	Reactive Orange 16	98-99	Lade et al (2015)

	Pseudomonas sp. SUK1	veratraldehyde, reducing O_2 to H_2O_2, which is used by peroxidases	Direct Red 81	99	Lade et al (2015)
	Comamonas sp. UVS		Red HE7B	57.5	Jadhav et al (2009)
Superoxide dismutase	*Lysinibacillus* sp.	Protect the cell from oxidative stress and have a role in decolorization along with oxidoreductive enzymes	Reactive Orange 16	100	Bedekar et al (2014)
Catalase	*Lysinibacillus* sp.		Reactive Green 19A	95	Bedekar et al (2014)

***Under Optimal Conditions**

FUTURE SCOPE

One of the shortcomings of microbial dye decolorization is the limited capacity of adsorption by the biomass. Moreover, the type of microbes to be used depends on the pre-treatment and properties of the effluents and their constituent dyes (Srinivasan & Viraraghavan, 2010). Screening of microbes that decolorize specific dyes is time consuming and laborious. Also maintaining purity of strains is difficult due to high chances of contamination (Bharagava et al, 2017). Furthermore, most microbes do not produce enough enzymes to decolorize huge batches of dyes. In such cases, genetic engineering is required to produce overexpressing strains. However, when compared to the huge list of advantages, these limitations can easily be overlooked, making bioremediation the solution of choice (Azubuike et al, 2016).

A lot of work still remains to be done to overcome the above limitations. Testing dye decolorization under high acidity and alkalinity needs to be studied, as most dyes render the pH of the effluent very high or low. Also, immobilizing the microbes may aid in reuse and easy separation of the biomass from the effluent. Studying various methods of immobilization and optimizing conditions for the same are necessary for sustainable dye decolorization (Sen et al, 2021b).

We also need to explore microbes that produce high yields of enzymes with unique and desirable properties, including resistance to extreme pH, temperature, dye toxicity etc. Thus, genetic engineering of such strains needs to be developed to produce strains that overexpress the dye decolorizing enzymes (Blumel et al, 2002), and also such that the enzymes are stable and resistant to extreme conditions. Furthermore, techniques on manipulation of enzyme activity need to be developed in order to obtain optimal decolorization.

CONCLUSION

In the present review, the role of all possible enzymes in biodegradation of dyes from the textile effluents were studied, along with the microbes that produce those enzymes, the dyes that are decolorized by them, and also the percentage of dye decolorization. It was observed that decolorization can be done by pure strains of bacteria, fungi, as well as consortia. It was also noted that most of the toxic azo and anthraquinone dyes were being decolorized by 80-100% under optimum conditions. Thus, it can be concluded that enzymes like azoreductase, NADH-DCIP Reductase, LiP, MnP, DyP, Laccase, Veratryl Alcohol Oxidase, Tyrosinase, Superoxide Dismutase and Catalase are indeed very efficient in decolorizing textile effluents with high concentrations of dyes, and demonstrated the possibilities of effective textile effluent treatment processes in the near future.

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