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"Gheorghe Asachi" Technical University of Iasi, Romania

LIGNINOLYTIC ENZYME SYSTEM OF WHITE-ROT FUNGI: A NATURAL APPROACH TO BIOREMEDIATION AND DETOXIFICATION OF AZO DYES IN TEXTILE WASTEWATER

Geetanjali Rajhans[1], Sudip Kumar Sen[2], Adyasa Barik[1], Sangeeta Raut[1]*

[1]Center for Biotechnology, School of Pharmaceutical Sciences, Siksha O Anusandhan (Deemed to be University), Bhubaneswar -751003, Odisha, India
[2]Biostadt India Limited, Waluj, Aurangabad-431136, Maharashtra, India

Abstract

In the wake of advance industrialization, the intensive growth of modern textile production and inappropriate wastewater treatment strategies have led to the release of noxious and carcinogenic contaminants like azo dyes directly or indirectly into the environment. Therefore, to ensure the protection of the humankind and natural bionetwork, cost-effective and efficiently regulated control measures are necessary. On this account, recent developments in biotechnology and microbiology have driven bioremediation of azo dyes using white rot fungi (WRF), which is a prospective option compared to conventional methods. These specially adopted microbes reductively cleave the azo group. This review has been carried out to address the bio-remedial capabilities of WRF in textile wastewater treatment by evaluating their typical attributes and performance. Furthermore, it emphasizes on the recent obstacles and future outlook for the abatement of azo dyes via advanced strains of WRF.

Keywords: azo dyes, bioremediation, detoxification, ligninolytic enzymes, textile effluent, white rot fungi

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1. Introduction

Over the past few years, owing to the threatening escalation of water pollution, the typical researches have started giving prime attention towards maintenance and management of water quality. The textile industries are adding to world's one-fifth of the industrial water contamination as they are one of the fastest growing industrial sectors on the globe (Lu, 2016). Although, pollutants may be emitted at each step of this industry chain, the processing operations like desizing, scouring, bleaching, dyeing, finishing and printing, possesses serious threats because of huge water demand as well as enormous wastewater production (Bhatia et al., 2017; Holkar et al., 2016). The effluent is excessively polluted with elevated BOD/COD ratio and total dissolved solids (Singare, 2019). Almost over 10,000 types of dyes and pigments are being used up by the textile industries, and a production of $7x10^5$ tons is annually produced (Ogugbue and Sawidis, 2011). Furthermore, it has been assessed that during the dyeing operations, 10-15% of dyes utilized in textile industries are being deposited in water effluents (da Silva Leite et al., 2016; Li et al., 2017).

Amongst the various dyes available for coloring cellulosic fibres, azo dyes are the broadest class of synthetic aromatic dyes which are stated to be the substantially predominant products (constituting >50% of all organic dyes produced) in the industrial effluents (Brüschweiler and Merlot, 2017). Azo dyes are synthetic organo-colorants that can be identified by the presence of one or more chromophoric azo (−N=N−) groups. The proclivity of textile industries

* Author to whom all correspondence should be addressed: e-mail: research.sangeeta@gmail.com; Phone: 9438450789

for azo dyes is owing to their remunerative attributes: easy and cost efficient synthetic process in contrast to the natural dyes, its effortless application, high molar extinction coefficient, high photolytic resistance, huge structural diversity, access to numerous radiant shades, strong cohesion to textile fibers and energy efficiency (Bafana et al., 2011; Brüschweiler and Merlot, 2017; Seesuriyachan et al., 2007). The lack of efficient elimination methods of azo dyes from the textile effluents may prompt contamination of the water resources. Azo dyes can be threatening to marine life by obstructing their normal photosynthetic processes (Lavanya et al., 2014) as well as terrestrial organisms including humans (Balakrishnan et al., 2016; Chequer et al., 2015; Du et al., 2015; Fernandes et al., 2015; Gadaleta et al., 2016; Ooka et al., 2016; Zanoni et al., 2013).

The azo dyes have harmful effects from their own action or aryl amine derivatives produced in the course of reductive biotransformation of azo bond. The chromophoric azo group (−N=N−) in the anionic and non-ionic dyes have been found to follow the course of reaction to undergo reductive cleavage (Xiang et al., 2016) (Eq. 1):

$$R{-}N{=}N{-}R{+}4e^{\mp}{+}4H{\rightarrow}R{-}NH_2{+}\,R'{-}NH_2 \quad (1)$$

For instance, if N-hydroxylamines are formed in the due course of azo bond cleavage, then it could quite possibly lead to DNA damage (Arlt, 2002; de Aragão Umbuzeiro et al., 2005). And because of their highly stable aromatic structures and strong tendencies towards aerobic conditions, the elimination rate of dyes at primary and secondary treatment phases of wastewater plants is insignificant. This creates easy carry-over of dye pollutants to water bodies, leading to bio-magnification in sediments and soil, resulting in contamination of ground water table (Salter-Blanc et al., 2016; Xiang et al., 2016). On this account, substantial research on both transitory and prolonged noxious aftermath of dyes on mankind and natural ecology, have been carried out (Long et al., 2017; Shabbir et al., 2017). In the attempt to prevent such threat, the surveillance and management of the acutely toxic and perilous dye pollutants is a major necessity.

Therefore, to decrease the pollutant generation, minimize fresh water consumption, and reuse of the effluent, it is essential to reprocess the wastewater containing dyes in a loop within the manufacturing unit (Ribeiro et al., 2017). On the above context, different physicochemical and biological techniques have been introduced by the researchers to remove dyes from textile wastewater (Morin-Crini et al., 2018; Yagub et al., 2014). But amongst all, biological method has been identified as a main prospective option for dyes removal, on the account of less sludge production, ecological viability, cost effectiveness and efficient detoxification of effluent (Ghosh et al., 2017; Ito et al., 2016).

In this treatment, microorganisms play a crucial role in mineralization of xenobiotic organic compounds. In addition, they have drawn interest towards textile dye remediation and detoxification because of their natural enzymatic mechanisms (Aljeboree et al., 2017; Durán and Esposito, 2000). The most implemented microorganisms are bacteria and fungi (Bafana et al., 2009; Chan et al., 2012; Fernando et al., 2014; Franciscon et al., 2009; He et al., 2004; Kalme et al., 2007; Kalyani et al., 2009; Nouren et al., 2017; Qu et al., 2012; Saratale et al., 2009; 2013; Tan et al., 2013, 2014, 2016; Wang et al., 2017). Even though, bacteria can eliminate azo dyes by breakdown of azo (−N=N−) linkage using specific azo-reductase, there is a high possibility for production of harmful and carcinogenic aromatic aryl amines as end products (Dawkar et al., 2009; Spadaro et al., 1992).

Furthermore, the degradation products when exposed to oxygen can quite possibly replace color to the effluent. In the other way around, fungi have (a) high tolerance to dye toxicity (Pinedo-Rivilla et al., 2009), (b) superior capacity to mineralize a wide range of persistent organo-pollutants such as azo dyes in both aqueous and non-aqueous medium (Okazaki et al., 2002; Selvam et al., 2003) and (c) capability to produce unspecific and non-stereoselective oxidative enzymes (Giardina et al., 2010; Wesenberg, 2003) that includes Laccase (Lac), lignin peroxidase (LiP), manganese peroxidase (MnP) (Giardina et al., 2010; Kuhad et al., 2004). Therefore, amongst the various types of microorganisms, fungi have been found to be competent in degrading and mineralizing the recalcitrant azo dyes (Rahimnejad et al., 2015).

Many experiments have been carried out for the degradation of dyes using whole cultures or fungal extracellular enzymes (Ghosh et al., 2018; Leonowicz et al., 2001). The fungi that have been observed to decolorize the wastewater are *P. chrysosporium, Trametes versicolor, Hirschioporus larincinus, Inonotus hispidus, Phlebia tremellosa, Aspergillus flavus*, etc. (Ghosh et al., 2018; Robinson et al., 2001).

A study says that *Aspergillus flavus* has been found to efficiently decolorize the wastewater comprising dye Acid brown 45 up to 75% within 50h (Ghosh et al., 2018). A few fungal strains, viz., *Candida* and *Magnusiomyces*, were found to completely detoxify and mineralize azo dyes (Brüschweiler and Merlot, 2017). A special class of fungi called White rot fungi has proven to be metabolically versatile in bio-remedial treatment of recalcitrant organo-pollutants such as azo dyes in the textile effluent. So far, these are the only organisms known to completely mineralize lignin to CO_2 and H_2O, however, they cannot use it as a sole carbon and energy source (Rekik et al., 2019). This study critically focuses on the potential bioremediation of azo dyes in the textile effluents by ligninolytic enzymes of white rot fungi.

2. White-rot fungi (WRF)

White rot fungi are very interesting fungal ecological group belonging to the class Basidiomycetes. Amongst the various class of fungi, they have the ability to digest the lignin component of lignocellulosic substrates (Dashtban et al., 2010). *Ganoderma lucidum, Phanerochaete chryrosporium, Trametes versicolor, Pleurotus ostreatus* and *Irpex lacteus* are few of the relevant species of WRF. Table 1 shows a report on the fungal cultures capable of dye degradation.

White rot fungi are so called because their extracellular enzymes produce white-colored cellulosic and hemicellulosic residues after degrading lignin (Arora and Sharma, 2009). WRF like *Phanerochaete chryrosporium, Trametes versicolor* and *P. avidoalba* are well known for decolorizing lignin-rich textile mill effluents as well as helping to reduce absorbable organic halides (AOX) and chemical oxygen demand (COD) of the effluent (Livernoche et al., 1983; Modi et al., 1998). It is also evident that the use of WRF is a suitable alternative for the bioremediation of dye-containing wastewaters from textile industry. The potential of these fungi is attributable to their biodegrading capability utilizing their ligninolytic enzyme system (mainly Laccase, lignin peroxidase, manganese peroxidase) (Ali, 2010). Other mechanisms such as biosorption and bioaccumulation could also be associated in removing dye by the fungal mycelia (Kaushik and Malik, 2009; Senthilkumar et al., 2014). White-rot fungus has the capability to breakdown azo dyes due to their structural similarity with lignin. The similar exceptional non-specific systems that enable these organisms to degrade lignin also facilitate degradation of the azo dyes. During the course of WRF metabolism, lignin oxidation by ligninolytic enzymes catalyzes the degradation/transformation of aromatic dyes in any of the two ways: (a) precipitation (b) opening the aromatic complex ring structure. Therefore, the fungus does not need any extra energy source (Husain, 2010). There are several variables that affect the decolorization using WRF. A recent study demonstrated that the most efficient nitrogen source for WRF is yeast extract, whereas the supplement of a carbon source was unnecessary to reach high levels of decolorization. They also showed that even though laccase production was favored by addition of copper, the decolorization rates were found to be unaffected (Merino et al., 2019). Another study worked on the parameters like carbon: nitrogen ratio, moisture content (%M), and copper sulphate concentration and found them to be inducers for ligninolytic enzymes (Jiménez et al., 2019). Further, Mejía and co-workers also demonstrated that a combined technology involving adsorption onto agro-industrial wastes and solid state fermentation using *Pleurotus ostreatus* can be a prospective option for best degradation (92.7%) of azo dyes like Allura Red, under the optimized media conditions such as carbon:nitrogen ratio of 2:1, moisture content of 80% and without $CuSO_4$ as inducer (Mejía et al., 2017).

The inadequate level of nutrients such as carbon, nitrogen sources frequently act as a stimulant for the synthesis and secretion of the ligninolytic enzymes (Ortiz-Monsalve et al., 2019). The laccase production can be enhanced by copper and numerous aromatic compounds in the medium. Previous study show that 5 mg l^{-1} of copper concentration seemed to be the optimal and coniferyl alcohol was found to be most effective inducing potential. New isoenzymes were formed after induction of each aromatic compound (Farnet et al., 1999). Another author demonstrate the enhancement in laccase activity in *Pleurotus ostreatus* by using wheat straw water extract as lignocellulytic enzymatic inducer (Parenti et al., 2013). While LiP activity in WRF can be significantly increased by using solid state fermentation (SSF) of lignocellulosic biomass such as jatropha, supplemented with the surfactant dodecyl sulfate (Ferreira da Silva et al., 2019).

Table 1. Examples of fungal strains capable of dye degradation

Fungi	***Dye***	***Decolorization (%)***	***Methodology implemented***	***References***
White rot fungi *Coriolopsis* sp. (1c3), isolated from compost source	Crystal Violet Methyl Violet Cotton Blue Malachite Green	94 97 91 52	*In vivo* (biodegradation)	Chen and Yien Ting. (2015a)
Aspergillus terreus GS28	Crystal Violet Methyl Violet Cotton Blue Malachite Green	95 98 82 54	*In vivo* (biosorption)	Chen and Yien Ting. (2015b)
Thielavia sp.	Remazol Brilliant Blue R	90	*In vitro* (enzymatic)	(Mtibaà et al. (2018)
Aspergillus terreus GS28	Direct Blue-1	98.4	*In vivo* (biosorption)	Singh and Dwivedi (2020)
White rot fungi consortium *(Daldinia concentric* and *Xylaria polymorpha*)	Cibacron brilliant red 3B-A	98	*In vivo* (enzymatic)	Bankole et al. (2018)

While Mishra and co-workers illustrated that ligninolytic enzyme activities were enhanced by metallic salts and phenolic compound supplements in SSF. Syringic acid supplemented medium increased the activities of LiP and lac, whereas gallic acid increased MnP activity and $CuSO_4$ increased Lac activity to improve the lignin degradation (Mishra et al., 2017). These attributes are supposed to play important role in design of process and optimization of fungal treatment of coloured effluents (Husain, 2010; Senthilkumar et al., 2014; Wesenberg, 2003).

2.1. Bioremediation capabilities of WRF

Different species of WRF have been shown to possess remarkable potential to bioremediate a wide range of dangerous organo-pollutants in industrial dye effluents, petroleum hydrocarbons, polychlorinated diphenyls, dioxins, pesticides, etc (Zhang et al., 2009). Their high tolerance to toxic environment, resistance to high temperature and a broad range of pH, makes them apt for bioremedial processes. The dye decolorization by ligninolytic enzymes of white rot fungi and estimation of ligninolytic activity of *Phanerochaete chrysosporium* was first reported by Glenn and co-workers (Glenn et al., 1983). On encounter, they adsorb dyes onto their hyphae, where they initiate the breakdown of the dye chemical bonds. The production of ligninolytic enzymes is highly conditioned by the composition of growth medium and culture conditions (Nerud and Mišurcová, 1996). Along with the lignin modifying enzymes (LME), WRF also produce and release redox mediators, which process electron movement and promote expansion of the substrate range for the ligninolytic enzymes (Cañas and Camarero, 2010; Marco-Urrea et al., 2010; Morozova et al., 2007). White rot fungi *Coriolopsis* sp (Chen and Yien Ting, 2015a), *Penicillium simplicissimum* (Chen and Yien Ting, 2015b) and *Pleurotus eryngii* (Hadibarata et al., 2013) showed degradation along with the COD removal.

Nevertheless, the cost of ligninolytic enzymes production has been a long-standing difficulty (Cardona et al., 2010). Biotechnological applications call for a huge amount of inexpensive enzymes. Therefore, the selection of appropriate lignocellulose biomass for fungal grow and production of enzyme is one of the crucial factors in development of competent biotechnology. Many studies have been carried out for efficient production of lignocellulolytic enzymes by WRF, which reveal that their production mostly depends on factors such as strain, substrate composition, ion concentration, and cultivation conditions (Buswell et al., 1995; Elisashvili et al., 2008; Stajić et al., 2006).

A more sophisticated solution to enhance ligninolytic activity is solid cultures or better known as Solid-State Fermentation (SSF). It is an optimal solution for cultivating fungi (Agosin and Odier, 1985; Tian et al., 2012; Wan and Li, 2010). Various studies have pointed that ligninolytic activity is more important with an SSF culture than with a liquid culture, perhaps because mechanisms are closer to those encountered in the natural environment. It is a fermentation process that involves a low water content of the substrate, with water/substrate ratios usually ranging between 1/1and1/10. By this process, oxygen diffusion and binding of enzymes to substrate is favored by the presence of mycelia, which is essential for fungal growth and leads to better lignin depolymerization. Lesser complex reactor designs than those of the liquid cultures, makes SSF a cheaper process (less aeration, mixing and heating). It also provides limited favorable environment for many microorganisms, and therefore lower sterilization energy costs. As a result, besides its ease of operation and cost-effectiveness, SSF with WRF would be advantageous compared to the enzymatic solutions. While modifying lignin, WRF increase its hydrophilicity (and thus its availability for hydrolysis).The mycelia penetration create spores, thus opening up a greater available surface area for enzymatic attacks.

With the use of SSF process, the natural habitat for most of the filamentous fungi can be maintained utilizing solid waste materials or inexpensive raw materials (Webb, 2017). Additionally, an SSF setting doesn't require the use of antifoam chemicals (Hölker and Lenz, 2005). Scale-up operations are hampered by the reduced control of online monitoring of process parameters, provision of heat and mass transfer as well as mixing (Singhania et al., 2009).

Another recently developed efficient process is submerged fermentation (SmF). It is the key method for production of enzymes including ligninolytic enzymes, owing to the effortless parameter control and better technological basis for scale-up to industrial trials (Singhania et al., 2010). This technology results in homogenous supply of nutrients which leads to full contact and nutrient adsorption by cultured microbes. Majority of filamentous fungi, including white rot fungi have a tendency to generate spherical pellets in a SmF setting, and this difference in morphology compared to SSF points towards a possible justification for the observed ligninolytic enzyme production. The setback observed with production of ligninolytic enzymes employing SmF is the multicellularity of white rot fungi, which hampers the cultivation productivity. Additionally, the mass transfer of oxygen can greatly affect the reproducibility of submerged cultivations.

A recent study reported the degradation of binary mixture of anionic dyes (brilliant blue FCF and allura red AC), using multiple WRF species under solid state fermentation (SSF) conditions. They found that *Irpex lacteus, Bjerkandera adusta* and *Trametes versicolor* achieved their maximum

decolorization of 80.11–86.04%, after 10-12 days. *I. lacteus* exhibited the highest decolorization percentage, even though only the enzyme manganese peroxidase was detected, with a maximum activity of 6.62 U gds^{-1} at day 14. Besides, *T. versicolor* was the only species with Lac activity, with a maximum of 15.94 U gds^{-1} at day 6 of fermentation (Merino et al., 2019).

Another study showed that *Trametes versicolor* under solid state fermentation conditions could degrade Red 40 dye adsorbed onto a low-cost waste product. Under the optimized conditions of carbon:nitrogen ratio (30:1), moisture percentage (75%), and inductor concentration (0.5 mM), maximum dye degradation of 96.04% was achieved. Also, the highest enzymatic activity was 8.49 U/gdm after 14 days of culture at the flask scale (Jaramillo et al., 2017).

However, free enzymes become unstable under certain harsh environmental conditions like temperature, pH, ionic strength of the solution, the type of solvent used, the amount and type of ions, inhibitors and cofactors present in the mixture, the concentration of substrates, the number of active enzyme molecules available during the catalytic conversion. Additionally, they are high-priced and non-reusable (Mohamad et al., 2015). Therefore, advanced strategies for stabilization of enzymes like immobilization procedures have been developed (Bilal et al., 2017).

Immobilization is a technique where the catalyst couple with an insoluble support matrix, to hold a proper geometry (Asgher et al., 2014). On the account of easy recovery from reaction mixture and handling convenience, immobilization provides stable catalysts for real-time applications. Besides, immobilization increases thermal stability and the enzymes become more resilient to degradation, denaturation, and aggregation (Bilal and Asgher, 2015). Many recent studies have reported the immobilization of ligninolytic enzymes using various strategies for their efficient industrial applications (Fernández-Fernández et al., 2013). Table 2 shows the immobilized ligninolytic enzymes from WRF and their effect on textile azo dye decolorization.

The above mentioned data reveals the potential utility of immobilization processes for onsite application of ligninolytic enzymes for better bioremediation of azo dyes.

3. Ligninolytic enzymes produced by WRF

White rot fungi produce and release lignin modifying enzymes and other lignin degrading compounds. Lignin modifying enzymes include laccase (Lac) (EC1.10.3.2), lignin peroxidase (LiP) (EC.1.11.10.14) and manganese peroxidase (MnP) (EC.1.11.113). The Lac and peroxidases differ mainly on the basis of their electron acceptor, the former use molecular oxygen (O_2) whilst the latter uses hydrogen peroxide (H_2O_2). In Table 3, the comparison between MnP, LiP and Lac from WRF is presented.

Due to difference in the redox-potential and extent of glycosylation, the catalytic potential and stability of ligninolytic enzymes vary. For enzyme-catalyzed reactions, the enzymes with high redox potential are favoured (Dashtban et al., 2010; Fabbrini et al., 2002; Riva, 2006). The redox-potential of ligninolytic enzymes is as follows: LiP>MnP> Lac. Glycosylation in extracellular enzymes can influence their shape, structure, composition, substrate binding sites formation and their properties such as enzymatic activity, redox-potential and catalytic potential (Sirim et al., 2011; Yang et al., 2015).

With increase in glycosylation, the enzyme stability increases but it may not always enhance enzyme's catalytic potential (Hamilton and Gerngross, 2007; Maestre-Reyna et al., 2015). It has been observed that deglycosylation of extracellular enzymes can have adverse effects on their stability, activity and catalytic potential (Nagai et al., 1997; Yang et al., 2015).

Table 2. Decolorization studies of textile azo dyes by immobilized ligninolytic enzymes of WRF

WRF	*Enzyme*	*Immobilization matrices*	*Immobilization technique*	*Dyes*	*Decolorization (%)*	*Time duration*	*References*
Aspergillus niger	Lac	Graphene oxide (GO)	Covalent attachment	DR 23 AB92	>75 >75	after 6 cycles	Kashefi et al. (2019)
Funalia trogii	Lac	Fe_3O_4-TCS hybrid composite	Cross-linking	RB 171 AB 74	>80 43	after 6 cycles 4th cycle	Ulu et al. (2020)
Ganoderma lucidum	MnP	Chitosan beads	Cross-linking	RB 21 RR 195A RY 145A	92.1 95.53 94.4	12h	Asgher et al. (2016)
Ganoderma lucidum	MnP	Agar-agar	Entrapment	RR 195A RB 21 RY 145A	78.6 87.4 81.2	12 h	Bilal et al. (2016)
Pleurotus ostreatus	Lac	Fe_3O_4/SiO_2 nanoparticles	Cross-linking	PR MX-5B	100	20 min	Dai et al. (2016)
Trametes versicolor	Lac	Copper alginate beads	Encapsulation	RBBR	75.8	4 h	Le et al. (2016)
Pleurotus ostreatus	LiP	Carbon nanotubes	Covalent attachment	RBBR	≥50	14 days	Oliveira et al. (2018)

Abbreviation: DR 23- Direct Red 23; AB 92- Acid Blue 92; RB 171- Reactive Blue 171; AB 74- Acid Blue 74; RBBR –Remazol Brilliant Blue R; RB 21- Reactive turquoise blue 21; RR 195A – Reactive red 195A; RY 145A- Reactive yellow 145 A; PR MX-5B- Procion Red MX-5B

Table 3. Comparison of the properties of MnP, LiP and Lac from WRF

E.C.	***Lac (1.10.3.2)***	***MnP (1.11.1.13)***	***LiP (1.11.1.14)***
	p-benzendiol: O_2-oxidoreductases	Mn(II): H_2O_2 oxidoreductases	diarylpropan O_2, H_2O_2 oxidoreductases
Prosthetic group	1 type-1-Cu, 1 type-2-Cu, 2 coupled type-3-Cu	heme	heme
MW (kDa)	59-110(tetramers≤390°)	32[a]– 62.5[b](122[a])	38-47
Glycosylation	N–	N–	N–
Isoforms	mono-, di-, tetramers; several	monomers; up to 11[d]	monomers; up to 15
pI	2.6–4.5	2.8[e] –7.2[f]	3.2– 4.7
pH range	2.0–8.5	2.6[g]– 4.5[h]	2.0– 5.0
E^0(mV)	500–800[k]	1510[i]	1450[j]
C–C cleavage	no	yes	yes
H_2O_2 –regulated	no	yes	yes
Stability	+ + +	+ + +	+
pIs of iso-enzymes[d]	2.6-4.5	2.9-7.0[c]	3.2-4.7
Natural mediator	3-HAA[n]	Mn^{2+}; Mn^{3+}	VA?[l], 2Cl-14DMB[m]
Specificity	broad, phenolics, incl. non-phenolics	Mn^{2+}	broad, aromatics
Secondary and synthetic mediators	ABTS[o], HBT[o], syringaldazine	thiols, unsaturated fatty acids	no

Modified from (Fakoussa and Hofrichter, 1999): [a]Basidiomycete strain RBS k1 (Willmann and Fakoussa, 1997); [b]Ceriporiopsissubvermispora in SSF (Lobos et al., 1994); [c](Thurston, 1994); [d]Ceriporiopsissubvermispora (Urzúa et al., 1995); [e]Nematolomafrowardii (Schneega et al., 1997); [f]Panaeolus sphinctrinus (Heinzkill et al., 1998); [g]P. tigrinus (Maltseva et al.,1991); [h]Pleurotusostreatus (Sarkar et al., 1997); [i]Chelator H_2O (Cui and Dolphin, 1990); [j](Schoemaker and Leisola, 1990) VA: Veratryl alcohol; [k](Messerschmidt, 1997); [l](Farrell et al., 1989; Tien and Kirk, 1983); [m]2Cl-14DMB:2-chloro-1,4-dimethoxybenzene (Teunissen et al., 1998); [n]3-HAA:3-hydroxyanthranilic acid (Eggert et al., 1995); [o]2,2V-ABTS: azinobis(3-ethylbenzthiazoline-6-sulfonate); HBT: 1-hydroxybenzotriazole (Bourbonnais et al., 1996)

LiP requires hydrogen peroxide (H_2O_2) to catalyze the oxidation of non-phenolic lignin units and mineralize the recalcitrant aromatic compounds. It has a high redox potential of 1.2V at pH 3.0 (Ertan et al., 2012) as compared with other peroxidases and does not require a mediator to oxidize phenolic and non-phenolic structures of lignin directly. Similar to LiPs, MnP also require H_2O_2 as an oxidant. MnP activity is mediated by manganese (Mn), where Mn^{2+} is oxidised to Mn^{3+}; eventually Mn^{3+} freely diffuses and gets involved as a redox couple in the oxidation reaction (Wariishi et al., 1989). It plays an important role in the initial stages of lignin degradation. In comparison to Lac, MnP leads to better degradation of phenolic lignin due to its higher redox potential (ten Have and Teunissen, 2001) with subsequent release of carbon dioxide (Morgenstern et al., 2008).

Initially, it was believed that Lac could only oxidize phenolic compound, because of its lower redox potential (450–800 mV) as compared to LiPs (>1 V). But in association with a mediator, a broad array of compounds can be oxidized by Lac. The mediators being low molecular weight compounds, transfer electrons from enzymes to substrate (Li et al., 1999).

Already, the enzyme characteristics, their mechanism of action as well as their biotechnological applications have been broadly depicted (Camarero et al., 1999; Hofrichter, 2002; Jones and Solomon, 2015; Ruiz-Duenas et al., 1999; Rodríguez Couto et al., 2006; Van Driessel and Christov, 2001). The ligninolytic enzymes in particular Lac and LiP have been observed to be very unambiguous in nature and distinctly efficient catalysts. It has been reported that these enzymes can catalyze the degradation and detoxification of a wide range of organo-pollutants like azo dyes present in industrial effluents (Bharagava et al., 2009, 2018; Mugdha and Usha, 2012; Pandey et al., 2007).

The data from the previous studies confirm the necessity of nutrient supplementation for more efficient colour reduction, owing to the fact that bioremediation is coupled with the production of ligninolytic enzymes in secondary metabolism (Swamy and Ramsay, 1999). For instance, the best biodecolorization of 93.8± 1.5% and 90.6±0.5% for Acid Red 357 and Acid orange 142, respectively were obtained when they were treated with reduced nutrient supply conditions. Although the treatment with high nutrient supply also showed efficient removal of colour, the biodecolorization efficacy was unexpectedly lower (90.0±0.5% for Acid red 357 and 84.5±1.0% for Acid orange 142). It was found that biodecolorization of real waste waters was highly influenced by composition of the nutrient sources (N_1– high nutrient source - 2% (m/v) of malt extract and 1% (m/v) of glucose; $N_{0.5}$ – reduced nutrient source-1% of malt extract and 0.5% of glucose; N_0- no nutrient), as biodecolorization is associated with ligninolytic enzyme production in secondary metabolism (Swamy and Ramsay, 1999). N_0 showed slight colour removal values, ranging between 50–70%. The coincidence of maximum peak of Lac activity and biodecolorization in case of $N_{0.5}$ and the delay in achieving higher biodecolorization rate in case of N_1 (compared to $N_{0.5}$) confirmed that a higher supply of nutrients can delay the biodecolorization/biodegradation of dyes. Moreover, N_1 permitted greater production of biomass, which seemed to negatively affect color removal.

The fungal morphology seemed to be associated with the better performance of reduced nutrient supply treatment. The treatment $N_{0.5}$ allowed to keep the uniform pellet form of the growing mycelia, homogeneously distributed in the

wastewater. In contrast, the high nutrient condition induced a heterogeneous growing of the inoculum, forming new pellets of different sizes that sometimes fragmented into small pieces or formed aggregates, resulting in a non-uniform mass of mycelium with small flakes peeling away. The pellet arrangement in the condition $N_{0.5}$ improved the mass transfer (oxygen and nutrients) from the liquid phase (culture medium) to the solid phase (growing cells) as reported by Kaushik and Malik (2009). Therefore, the reduced nutrient supply treatment could help achieve better results because of better physiological conditions that led to high peaks of Lac activity (1000–1300 U L^{-1}) and mycelia pellet arrangements (Ortiz-Monsalve et al., 2019).

Another study showed that lignocellulolytic enzyme production by the WRF, *Pleurotus ostreatus* could be improved by using different lignocellulosic biomasses as a substrate in sequential SSF and SmF processes. A higher yield of Lac (543 ± 21 U/L) was achieved. The results showed that the fermentation method and nature of the lignocellulosic biomass have important role in lignocellulolytic enzyme expression. This indication would be helpful in optimizing the production of integrated industrial lignocellulolytic enzymes (An et al., 2016).

Fang et al. (2018) showed that the WRF strains *Trametes Versicolor* (strain MES 11914) and *Pleurotus Sajor Caju* (strain MES 03464) could be grown using solid-state fermentation of solid digestate and enhance the secretion of ligninolytic enzymes such as Lac and MnP to degrade lignin in different extents (Fang et al., 2018).

3.1. Laccase

Laccases (oxygen oxidoreductase) are N-glycosylated multi-copper proteins with molecular masses of 60-390 kDa and are efficiently produced by a wide spectrum of Basidiomycota (Matera et al., 2008; Nguyen et al., 2016; Songulashvili et al., 2016; Surwase et al., 2016). Lac was first discovered in the sap of the Japanese lacquer tree *Rhus vernicifera*, and its characteristic as a metal containing oxidase was discovered by Bertrand in 1985 (Giardina et al., 2010) and the active site is occupied by four copper atoms (as Cu^{2+} in the resting enzyme) distributed among different binding sites (McGuirl and Dooley, 1999; Messerschmidt, 1997; Wesenberg, 2003). Their low substrate specificity has helped them draw tremendous attention in environmental, industrial and biotechnological sectors (Agrawal et al., 2018).These are oxidative extracellular enzymes synthesized by white rot fungi and are proficient in degrading different types of lignin-based compounds *in vitro.* Because of their capacity for bioremediation and distinctive features such as non-specific oxidation capacity, no requirement for co-factors and no dependence on readily available oxygen as an electron acceptor, Lacs have great importance in different biotechnological processes (Kalyani et al., 2012; Telke et al., 2011). They can oxidize phenols and phenolic lignin compounds by one electron abstraction resulting in the formation of radicals that can either repolymerize or lead to depolymerization (Demarche et al., 2012; Surwase et al., 2016). A number of studies have published on Lac mediated degradation of azo dyes (Balan et al., 2012; Palvannan and Sathishkumar, 2010; Sathishkumar et al., 2013). Table 4 shows various Lac producing fungal cultures and their ability to degrade numerous azo dyes. Lac acts by formation of free radicals which bypass the steps involved in the formation of carcinogenic amines (Chivukula and Renganathan, 1995). White rot fungi can easily enhance Lac production by addition of inducers (Palvannan and Sathishkumar, 2010). The mediators being low molecular weight compounds which carry electrons from enzymes to substrate. The mediator can easily access the active site of the enzyme, where it gets oxidized into more stable intermediate (high redox potential).

Table 4. Decolorization of various azo dyes by Lac producing fungal culture

S. no.	***Fungal culture***	***Dye***	***Time***	***Decolorization (%)***	***References***
1	*Cerrena unicolor*	Acid Red 27	24 h	100	Michniewicz et al. (2008)
2	*Geobacillus catenulatus MS5*	Congo Red	32 h	99	Verma and Shirkot (2014)
3	*Pleurotus ostreatus*	Synazol Red HF6BN	24 days	96	Ilyas et al. (2012)
4	Immobilized *Trametes pubescens*, *Pleurotus ostreatus*	Remazol Brilliant Blue R, Reactive Blue 49	10 days	>95	Casieri et al. (2008)
5	*Ganoderma* sp.	Methyl Orange	72 h	>90	Zhao et al. (2011)
6	*Armillaria* sp. F022	Reactive Black 5	96 h	80	Hadibarata et al. (2012)
7	*Pleurotus ostreatus*	Remazol Brilliant Blue R	72 h	80	Palmieri et al. (2000)
8	*Lentinus Polychrous*	Congo Red	3 h	75	Suwannawong et al. (2010)
9	*Pycnoporus sanguineus*	Trypan Blue	24 h	70	Annuar et al. (2009)
10	*Coprinopsis cineria*	Methyl Orange	4 h	47.60	Tian et al. (2014)

After diffusing away from the enzyme, the oxidized mediator oxidizes more complex substrates before returning to its original state (Barreca et al., 2003; Bourbonnais et al., 1995; Eggert et al., 1996; Fabbrini et al., 2002; Fernández-Sánchez et al., 2002; Johannes and Majcherczyk, 2000; Shleev et al., 2005; Solomon et al., 1996; Xu, 1997; Xu et al., 1999). The electrons taken by Lacs are finally transferred back to oxygen to form hydrogen peroxide. Enzymes are mostly substrate specific, but Lac can oxidize a wide range of substrates like aromatic amines, diphenols, polyphenols, benzenethiol. The ideal redox mediator would be a small-size compound, able to generate stable radicals (in its oxidized form) that do not inactivate the enzyme, and whose reactivity would allow its recycling without degeneration. In addition, from the point of view of their industrial and environmental application, Lac mediators should be environmental-friendly and available at low cost. The most competent Lac mediators for oxidation of recalcitrant aromatic compounds are 2, 2-azino-bis-3-ethylbenzothiazoline-6-sulfonic acid (ABTS) and the NN–OH mediators, such as hydroxyphthalimide (HPI), 1-hydroxybenzotriazole (HBT), N-hydroxyacetanilide (NHA) or N-violuric acid (VLA) (Call, 1994; Paice et al., 1995; Srebotnik and Hammel, 2000; Xu et al., 2000). When Lac oxidixes these mediators, a highly reactive nitroxyl radical (NN–O·) is generated due to the release of a proton after enzymatic removal of an electron. The target substrate is then oxidized by the nitroxyl radicals by the mechanism of hydrogen atom transfer (HAT) (Fabbrini et al., 2002; Xu et al., 2000).

Lacs have been extensively studied in huge scale for their capability to degrade azo dyes (Casieri et al., 2008)(Blánquez et al., 2004; Chivukula and Renganathan, 1995; Kirby et al., 2000; Novotný et al., 2011; Peralta-Zamora et al., 2003). On supplementation of Cu^{2+} (Palmieri et al., 2000) or aromatic compounds such as veratryl alcohol (Rodriguez-Couto et al., 2006) and 2,5-xylidine (Eggert et al., 1997; Leonowicz et al., 2001), Lac production is enhanced. Lac alter the structure of azo dye by destroying their chromophoric assemblies, with the generation of phenoxyl radicals in the course of reaction (Chivukula and Renganathan, 1995). During the first step, a phenoxy radical is generated after an electron is abstracted from the phenolic/naptholic ring. The abstraction of a second electron yields an aromatic cation which can be stabilized by the electron-donating groups present in the ring (Fig. 1).

Previously, the best biochemical decolorizations were achieved with those azo dyes that carried hydroxyl (–OH) functional groups (that are strong electron donating moieties) in ortho and para positions to the azo bond (Kandelbauer et al., 2004a). The Lac alone cannot attack the meta-substituted analogue because of less activation energy at this position. The electron withdrawing substituent like halogen or nitro groups on the aromatic rings, make it difficult for the oxidases to yield cation radicals, which inhibits the dye degradation. Alternatively, the azo dyes characterized by weakly electron-donating methyl groups have been observed to decolorize efficiently (Pasti-Grigsby et al., 1992). And the heterocyclic azo dyes containing pyrazole or triazole rings were not significantly decolorized unless there are hydroxyl and other electron donating groups present on the heterocyclic and vicinal aromatic rings in the ortho position to azo bond. Other effects may contribute as well such as those caused by reaction intermediates (Kandelbauer et al., 2004b).

The production of Lac is dependent on the nature of carbon source, which might come from various agro-industrial lignocellulosic residues. But there are limitations to Lac production due to lack of kinetic and design data related to several fermentation processes (Lonsane et al., 1985). SmF can enhance Lac production in a comparatively short period than SSF (Songulashvili et al., 2007). The physiological modulation of Lac production is relatively simpler in SmF than in SSF (Elisashviliet al., 2008).

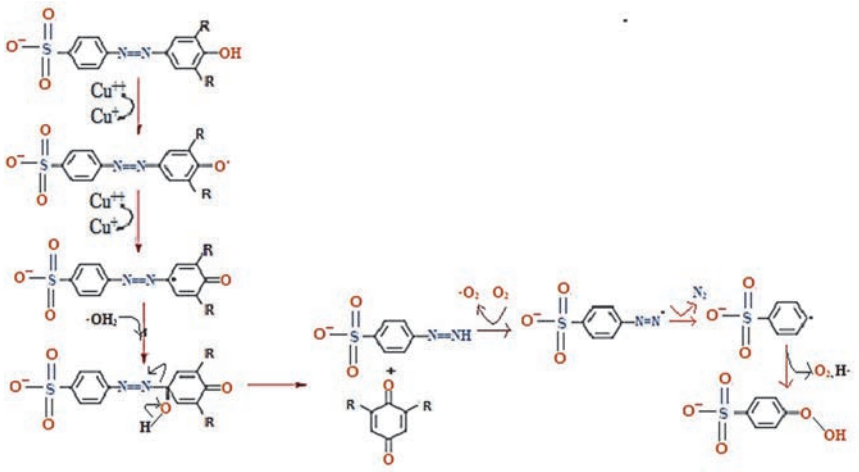


Fig. 1. Proposed mechanism for the degradation of phenolic azo dyes by *P. oryzae* Lac (Chivukula et al., 1995)

Lac production by WRF can be successfully carried out in SmF, by substituting the carbon source with various agro-industrial lignocellulosic residues (Songulashvili et al., 2011). This approach would trim down the production cost of ligninolytic enzymes and hence allow large-scale industrial applications. A previous study demonstrated that Lac production by the WRF *Cerrena unicolor* C-139 can be enhanced by SmF in the presence of a cheap lignocellulosic substrate, wheat bran. A maximum Lac activity of 416.4 U mL^{-1} at day 12 of fermentation was observed (Songulashvili et al., 2016).

The degradation mechanism of crystal violet dye by Lac with a a low molecular mass fraction (LMMF) extracted from WRF *Pleurotus ostreatus* has been reported by Yen and co-workers (Yan et al., 2009). Other author showed that on Lac assisted degradation, the dye azonaphthol Orange 2 was found to have 72.8% decolorization, whereas the dye azobenzene Acid Orange 6 had 45.3% decolorization (Legerská et al., 2018). The presence of hydroxyl group at o-position to azo bond in the structure of Orange 2 was more favoured than the presence of two hydroxyl groups at o- and p-positions to azo bond in Acid Orange 6. Even though the Lac treatment was more efficient in case of Orange 2 decolorization, the toxicity of both the monoazo dye solutions was lessened for the prokaryotic growth. Their result suggested that *T. versicolor* derived Lac has the capability to degrade selected synthetic dyes by decreasing the toxic effect of synthetic dyes after Lac-catalysed reaction.

However, it is also crucial to research engineering aspects for industrial applications of Lacs. Certain pilot scale bioprocesses have also been put to action for Lac production. Some authors demonstrated a cost-effective process for higher production of extracellular, thermo-alkali stable Lac from *Staphylococcus arlettae* S1-20 using tea waste. And a pilot-scale bioprocess with optimal conditions increased Lac yield.

The optimum temperature (85ºC) and pH (9.0) retained significant amount of activity even in the presence of 20% (v/v) ionic liquids (Chauhan et al., 2018). Studies on bioreactor scale-Lac production reported that the Lac production (3.80 U mL^{-1}) by the fungus *Pleurotus ostreatus* CP-50 in a 10 L stirred tank bioreactor was enhanced in low oxygen transfer rates (Tinoco-Valencia et al., 2014). Another report shows that a marine-derived basidiomycete *Peniophora sp.* CBMAI 1063 could produce considerable amount of enzyme in both stirred tank (ST) and air-lift (AL) bioreactors. ST bioreactor led to higher Lac production, while the AL bioreactor supported higher formation of biomass. They revealed that initial pH of the medium, agitation and aeration rates, directly influences Lac production and fungal biomass formation (Mainardi et al., 2018).

3.2. Lignin peroxidase

Lignin peroxidase (LiP) (1,2-bis(3,4-dimethoxyphenyl)propane-1,3-diol) also known as ligninase is a key-lignin degrading enzyme produced by white rot fungi. They have molecular weight ranging from 41-43 kDa. In presence of hydrogen peroxide, LiP carry out the oxidative cleavage/depolymerization of lignin. LiP was primarily isolated from the culture broth of a ligninolytic fungus, *Phanerochaete chrysosporium*. LiPs are oxidized by H_2O_2 to give a two-electron-oxidized intermediate (LiP-I), where iron is present in Fe^{4+} state and a free radical is found on the tetrapyrrole ring or on a nearby amino acid. Subsequently, LiP-I oxidizes a donor substrate by one electron which produces a radical cation and LiP-II, where iron is present in Fe^{4+} state, but no radical is found on the tetrapyrrole. Then a second molecule of donor substrate is oxidized by LiP-II, yielding another radical ion and the resting state of peroxidase. Non-phenolic units of lignin are oxidized by LiP via removal of an electron and creating cation radicals, which then decomposes chemically. The Cα-Cβ bond in the lignin molecule is cleaved by LiP (Hatakka, 2005; Wong, 2009). The general catalytic mechanism followed by LiP (Wong, 2009) (Eqs. 2-4):

$$LiP(Fe^{3+}) + H_2O_2 \rightarrow LiP - I(Fe^{3+}) + H_2O \quad (2)$$

$$LiP - I + AH \rightarrow LiP - II(Fe^{4+})A \quad (3)$$

$$LiP - II + AH \rightarrow LiP\ A \quad (4)$$

LiP breaks down lignin in an approach similar to that of Lac, MnP and several other peroxidases such as versatile peroxidase (Zeng et al., 2013).

Accounting to its low specificity and high redox potential, LiPs have been characterized by a distinct ability to oxidize a vast range of aromatic phenolic and non-phenolic compounds as well as organo-pollutants like azo dyes (Valli et al., 1990). Recently LiP obtained from *Ganodermalucidum* IBL-05 showed decolorization efficiency for Sandal-fix Red C4BLN, Sandal-fix Turq Blue GWF, Sandal-fix Foron Blue E2BLN, Sandal-fix Black CKF and Sandal-fix Golden yellow CRL dyes of 66%, 59%, 52%, 40% and 48% respectively, which then significantly increased to 93%, 83%, 89%, 70% and 80% in case of Ca-alginate immobilization of LiP (Bilal et al., 2019). It can be concluded that immobilized LiP might be a potential biocatalyst for the bioremediation of dye-based textile effluents. Fig. 2 shows a schematic degradation pathway of methyl orange, as a model dye, in the presence of LiP (Bilal et al., 2018).

3.3. Manganese peroxidase

Amongst the various ligninolytic enzymes, manganese peroxidase (MnP), a heme protein with molecular weight normally varying from 40 to 50 kDa (Hofrichter, 2002), are thought to play a crucial role in lignin breakdown because it is found in all lignin degrading WRF. They belong to the class II peroxidase group in Basidiomycetes fungi and possess a highly specific Mn^{2+} binding site. MnP was first extracted in the culture extract of *P. chrysosporium* (Bonnarme and Jeffries, 1990).

The classical long MnPs have three amino acid residues (one Asp and two Glu) in their binding site while several fungal Mn^{2+} oxidizing enzymes have been found with an additional tryptophan residue on the enzyme surface, which are called hybrid MnPs. These hybrid MnPs resemble with LiPs and can perform oxidation through a long range electron transfers. MnP has been found to decolorize majority of sulfophthale in dyes at pH 4.0.

Previous research have deduced that MnP activity has strong preference for methyl group at ortho than at the meta position on chromophore, as MnP has higher Km value for meta-cresol purple and lower Km value for ortho-cresol red (Shrivastava et al., 2005). The mechanism of the catalytic activity of MnP is as follows (Isroi et al., 2011; Wong, 2009) (Eqs. 5-8):

$$MnP + H_2O_2 \rightarrow MnP - I + H_2O_2 \quad (5)$$

$$MnP - I + Mn^{2+} \rightarrow MnP - II + Mn^{3+} \quad (6)$$

$$MnP - II + Mn^{2+} \rightarrow MnP + Mn^{3+} + H_2O \quad (7)$$

$$Mn^{3+}RH \rightarrow Mn^{2+} + RH \quad (8)$$

where: *RH* = organic substrate.

MnP in crude form has the capability to decolorize dyes like indigo carmine (Li et al., 2015). Studies have shown successful decolorization and removal of organic matter of Acid Red 88 (dye removal efficacy of 96%) and Reactive Red 180 (dye removal efficacy of 98%) with the help of enzymesfrom WRF *Phanerochaete chrysosporium* in a bioreactor system (Deveci et al., 2016).

Fig. 3 shows a peroxidase-assisted degradative pathway of Reactive black 5 and Reactive Black 19. Another research demonstrated the decolorization of azo dyes like amaranth, reactive black 5 and cibacron brilliant yellow to up to 95, 76 and 46% respectively, up to 24 hours by the action of a purified MnP (MnP TP55) whereas the decolorization efficacy was found to be 90.55 and 88% respectively on the action of another purified MnP (MnPBA30) (Rekik et al., 2019). Both the MnPs were isolated from a WRF *Trametes pubescens* strain i8. Certain studies on decolorization by purified MnPs show that MnP have requirement of H_2O_2 and $MnSO_4$ to attain maximal rates of decolorization (Champagne and Ramsay, 2005).

4. Elucidation of degradative pathways employed by WRF in decolorization of azo dyes

Dye removal mechanism by WRF follow three major steps i.e., bioaccumulation, bio-absorption and biodegradation. Usually the actively growing microbes carryout the bioaccumulation process as a result of their metabolism, while biosorption is found to occur in both living and dead biomass. Biodegradation takes place as a result of breakdown of dye molecules by the naturally produced versatile extracellular and intracellular enzyme activities of the fungi such as Lac, MnP and LiP (Tan et al., 2016). Fungi carry a superfamily of intracellular heme-containing monooxygenase called cytochrome P450s (CYP), which have crucial role in housekeeping biochemical reactions, detoxification of xenobiotics and sustainability in adverse ecological niche (Durairaj et al., 2016) (Fig. 4).

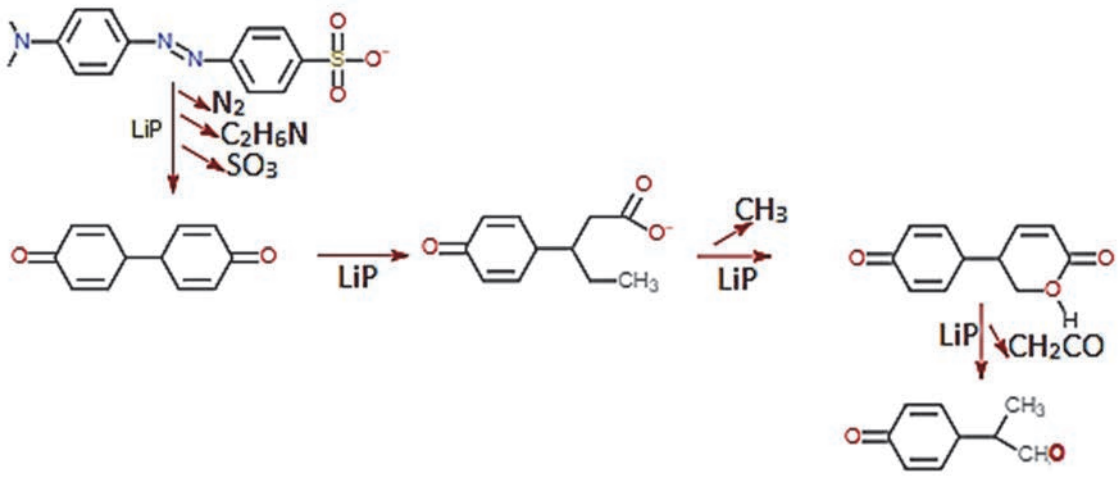
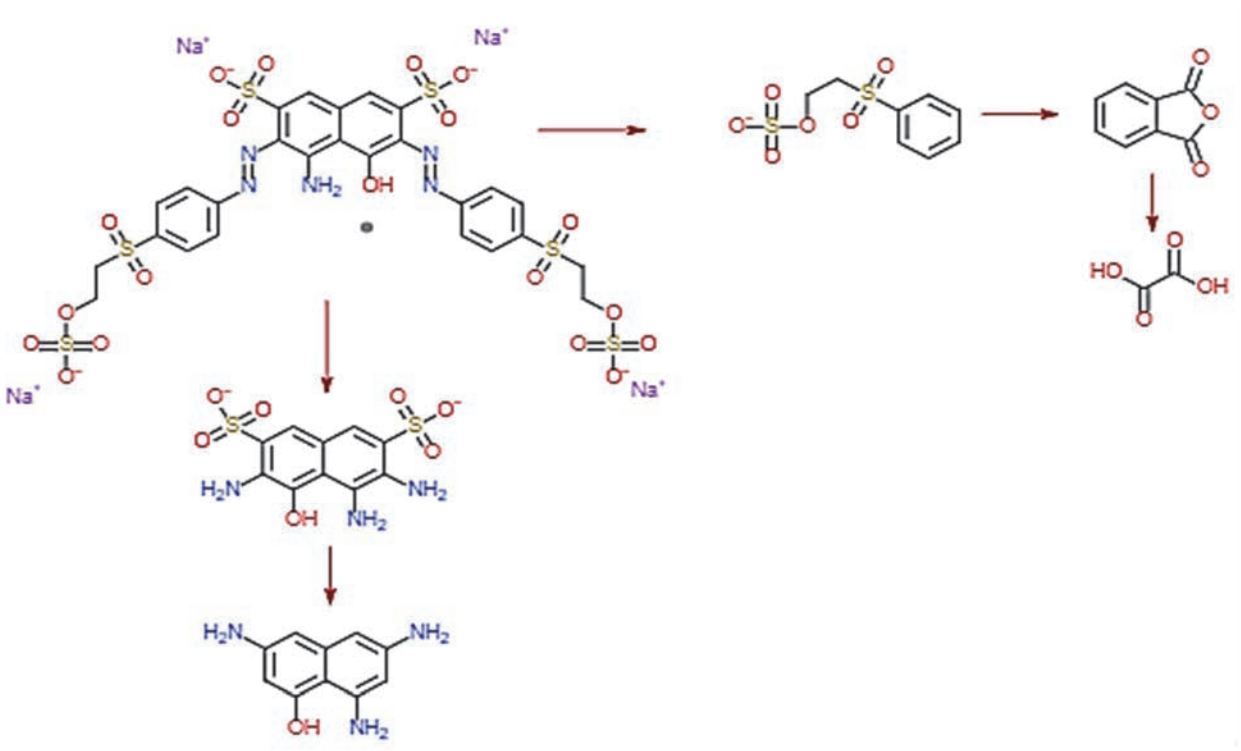
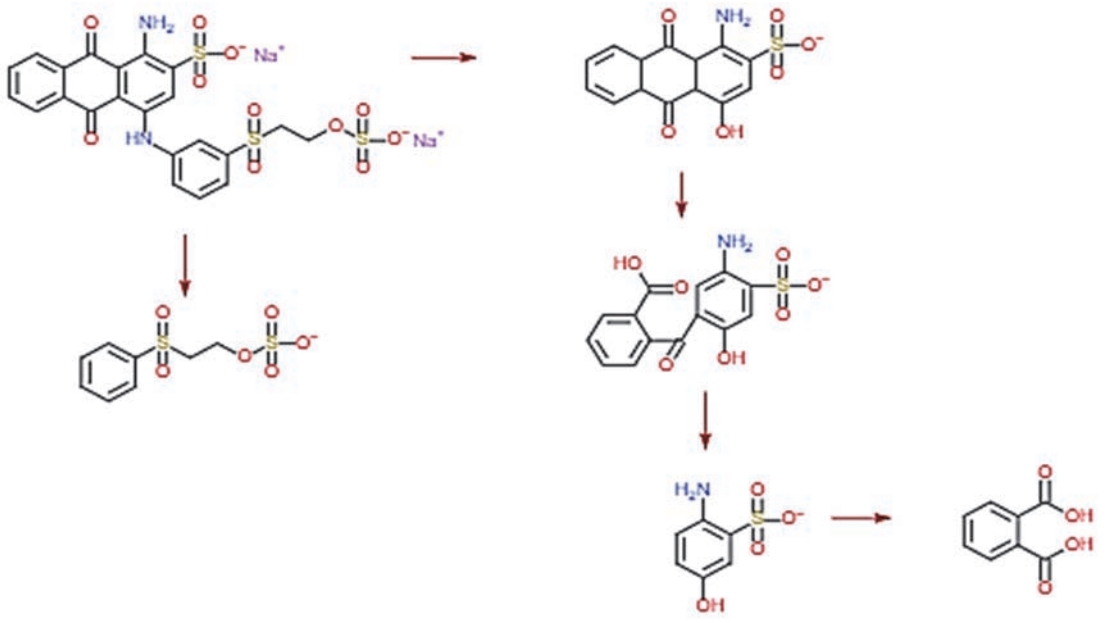


Fig. 2. A schematic degradation pathway of Methyl Orange, as a model dye, in the presence of LiP as a novel catalyzing agent (adapted from Bilal et al., 2018)



(a)



(b)

Fig. 3. Peroxidase-assisted catalytic pathways; (a) RB-5, and (b) RB-19 (adapted from Bilal et al., 2018)

Over 6000 CYP genes are found in fungal genomes (Manavalan et al., 2015). The presence of lignin-based substrates provided by Malt Extract Agar (MEA) media, leads to activation of these fungal enzymes (Ghorbani et al., 2015; van Kuijk et al., 2015). The cytochemicals mediated by the CYP enzyme system can transform the dye molecules into chemical derivatives such as hydroxyl, dihydrodiol and quinone.

The subsequent step is thought to be reductive breakdown of azo groups in the dye molecules by oxidoreductases. Thereafter, these metabolites get coupled with other functional groups such as methyl and glucose groups mediated by transferases, leading to de-aminated dye molecules (partly degraded) and formation of simplest end products (Manavalan T. et al., 2015).

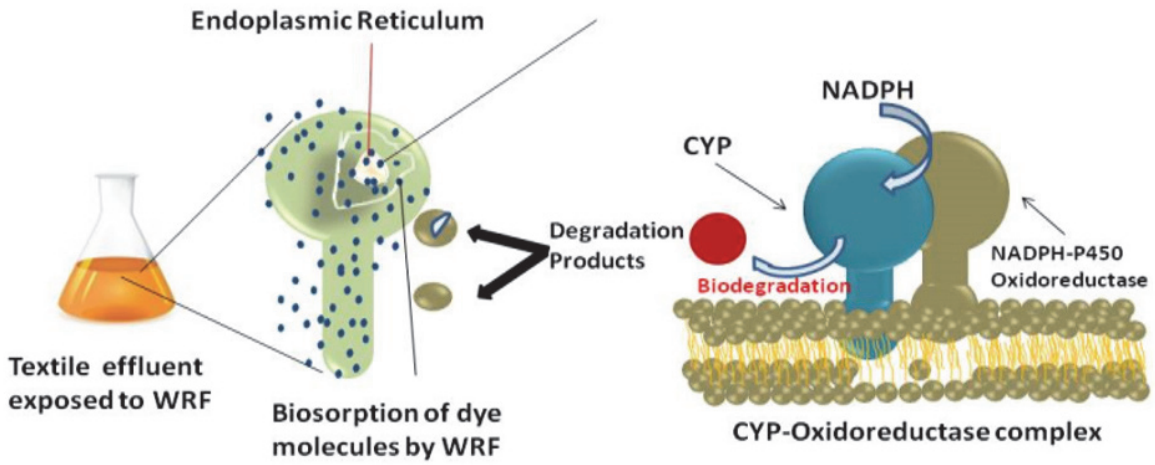


Fig. 4. Proposed WRF mechanism for dye degradation (adapted from Kumar et al., 2018)

Then the degraded products can either get stored in the cell organelles or get released into the environment, where they may be further broken down by surrounding media and /or organisms. Previous studies depict this mechanism and thus, it is essential to thoroughly investigate on the same.

5. Challenges and future scope of WRF for dye remediation

Extensive studies to biodegrade and detoxify azo dyes in textile effluents using various WRF strains have been followed through. These treatments have been found to be a propitious option. The analysis of operational performance reveals that for proper maintenance of the system, the microbe-based dye degradation is still unfortunately reliant upon the alteration in the environment of the microbial population. Additionally, the trouble-free access to dye molecules by microbes is another important factor that would regulate the operational success. To overcome this issue by ligninolytic enzymes in the bioremediation of contaminants in situ, immobilization should be extensively implemented in order to improve stability, adaptability and commercial feasibility of enzymes. The best method will also be scrutinized by the principal parameters like appropriate transfer of oxygen, less operation time, homogenization, operational stability and the suitability in scale up. In the advancement of this microtechnology towards industrial scale, sterility must be avoided. Since the wastewater sterilization is not feasible from an economic and environmental point of view, microtechnology should emphasize on non-sterile conditions. This approach would assure endurance and activity of the microbes during biodegradation operations. Besides, the vital issues thwarting the huge-scale biodegrading approach of WRF is the requirement of immense amount of ligninolytic enzymes and the accompanying excessive production cost. The use of SmF or SSF technologies can be implemented to improve the low cost production of ligninolytic enzymes by utilizing cheaper lignocellulosic biomasses as the inducer substrate. Also, new strategies need to be developed to curb the costs that mainly account to the growth media for the fungi during the production of ligninolytic enzymes. Moreover, the reported experiences in pilot plant are still too inadequate. Therefore, before a full-scale application, it should be essential to execute the outcomes inferred from bench-scale reactors to the pilot plants. Future research should be based on:

a. Application of a standardized setup of parameters for determining whether SmF or SSF is the optimal cultivation process for specific strains of WRF and selection of appropriate substrates like agro-industrial residues, through inter- and intra-laboratory trials. The selection of the appropriate processes is of great importance with respect to optimized enzyme-product yield and shaping future researches on solid state or submerged fermentation technologies.

b. The nature of the lignocellulosic biomass and the fermentation method play an important role in lignocellulolytic enzyme expression. This hint would be supportive in optimizing the production of integrated industrial lignocellulolytic enzymes.

c. Immobilization of ligninolytic enzymes should address an existing issue such as suitability of unique physiochemical and structural features of an enzyme for bioremediation of azo dyes at a larger scale. Furthermore, the bioremediation carried out using immobilized ligninolytic enzymes should be eco-friendly and cost effective. On the account of being environmentally friendly, non-toxicity and ease of use, the development and implementation of immobilized enzymes are supposed to be an area of intense future investigations.

d. Intensive studies on the operational parameters for the dye biodegradation, so as to enhance the efficacy of microbial system towards the breakdown of the dyes.

e. In order to surmount technical challenges and escalate the feasibility of biodegradation

activities on the basis of kinetics, stability and operational capabilities.

f. Results derived in bench-scale reactors need to be substantiated at pilot plants in real time under real reaction conditions (pH, temperature, etc) before any full-scale application.

6. Conclusions

The progress and execution of microtechnology's for environmental management is a need for sustainability. So far, assorted physicochemical treatment strategies have been brought to action to curtail the overall degree of dye pollution in the aqueous ecosystem.

Nonetheless, the effectuality of these conventional methods is limited on account of high operating/ energy costs, enormous sludge production, release of environmentally unfriendly byproducts and need for huge amount of chemicals and attracting energy penalties.

This review unfolds that the white rot fungi are the most promising organisms with potential uses in biodegradation and management of recalcitrant environmental contaminants and xenobiotics like azo dyes. With the combined use of prospective technologies like SSF and SmF, the productivity and activity of ligninolytic enzymes can be enhanced by many folds.

The commercial and onsite application efficiency of WRF and its ligninolytic enzymes can be enhanced by immobilization techniques. In conclusion, the WRF could be envisioned as an outstanding alternative for bioremediation and detoxification of textile wastewater, as they have been recognized as advantageous for dye removal on the grounds of lucrative operations, eco-friendly approach, effortless, safe operations and zero sludge production.

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