Type of the Paper (Review)

Biological Consortia Designed for Laccase Production and Dye Removal

Roberto González [1], Roberto Villagómez [1], Alfredo Madariaga[2], Javier Castro [1] and Cesar González, [3]*

[1] Academic Area of Chemistry, Institute of Basic Sciences and Engineering, Autonomous University of Hidalgo State (UAEH)-City of Knowledge, Pachuca-Tulancingo Highway Km 4.5, Col. Carboneras, Mineral de la Reforma, Hidalgo, C.P. 42184, Mexico; zerj_000@hotmail.com (R.G.); jrvi@uaeh.edu.mx (R. V.); jcastro@uaeh.edu.mx (J.C.).

[2] Institute of Agricultural Sciences, Autonomous University of Hidalgo State, Rancho Universitario. Av. Universitaria Km 1, Ex. Hda. Aquetzalpa AP 32, Tulancingo, Hidalgo, C.P. 43600, Mexico; alfredomadariaga60@gmail.com (A.M.).

[3] *Apan-Calpulalpan s/n Highway, Colonia Chimalpa Tlalayote, Apan, Hidalgo, C.P. 43920, Mexico, correspondence: ccr_gonzalez@yahoo.com (C.A.); Tel.: (+52 771) 717 2000 Ext. 5800, 5801.

Abstract: The potential of biological consortia designed for laccase production and dye treatment is discussed in this review. The poor yields in laccase production and low efficiency in dye decolorization of monoculture-based treatments has encouraged the use of designed biological consortia. A consortium is a system where the growth of two or more organisms, chosen to improve a particular bioprocess, is induced in the same medium. Chemical and natural mediators are being replaced by consortia for the production of laccases because, in addition to being less toxic, they induce new enzyme isoforms and lead to high laccase yields. On the other hand, consortia act synergistically in the decolorization of azo dyes through the enzymes they produce, so overall degradation is improved. Designed consortia are an attractive alternative still in development that could provide new biotechnological tools for the treatment of textile effluents.

Keywords: designed consortium; textile effluents; biological induction; azo dyes

1. Introduction

The enforcement of increasingly stringent regulations on the treatment of textile effluents has boosted the development of biotechnological approaches for waste treatment and remediation [1, 2], especially because biotechnological tools base their action on cellular enzymes, and thus are considered as "environmentally friendly" technologies. These biotechnological tools are based on vegetables [3], lichens [4], algae [5], bacteria [6], filamentous fungi [6], bacterial consortia [7, 8], and designed consortia [9]. A common trait in these organisms is the production of laccases, an enzyme class capable of degrading dyes. Laccase production depends on chemical inducers [10] or natural inducers such as biological consortia [11]. Several studies have shown that consortia allow higher laccase yields than monocultures [12], with the additional advantage of generating different enzyme isoforms [13]. In dye treatment studies, biological consortia have increased dye removal rates [14] because they are less susceptible to organic contamination [15] and exhibit higher enzyme production levels [16, 17], increased resistance to abiotic conditions [18], and less enzyme inactivation rates [19]. The first type of consortia studied for laccase production and dye removal were naturally occurring ones, in which microorganisms were extracted from contaminated sites; despite their efficiency, their growth and properties as degraders are difficult to control [20], so their application is limited. On the other hand, designed consortia are systems where the growth of two or more organisms is induced in the same medium; while the organisms are specifically chosen to improve a given bioprocess, there are few reports in the literature about the use of consortia for the production of laccases and the removal of dyes in waste waters. Therefore, the role of designed consortia in laccase production and dye removal is discussed in this review as a biotechnological tool to reduce the contamination caused by textile effluents.

2. Biotechnological methods for treatment of textile waste water

2.1. Pollution by textile effluents

The textile industry negatively impacts on the environment due to the toxicity of its effluents [21]. It is estimated that discharged waste waters have a COD (chemical oxygen demand) of 115-175 kg per ton of finished textile [22]. The dyeing and finishing processes are the primary sources of contamination in textile waste water [23].

It has been reported that 80 000 tons of dyes are discharged in natural water bodies per year worldwide, and 60% to 70% of those are azo dyes [24]; thus, azo dyes are major contaminants in textile effluents. Azo dyes inhibit aquatic photosynthesis, reduce dissolved oxygen, and are toxic to flora, fauna, and humans [25]. Some azo dyes are category 3 carcinogens, according to the International Agency for Research on Cancer [26]. During water treatment azo dyes are transformed into toxic compounds, including sulfonated aromatic amines, phenol, and naphthalene [8].

The European [1] and North American [2] legislation requires that effluents of the textile industry are treated, avoiding the discharge of compounds that are harmful for human health and the environment as much as possible. A brief description of the physicochemical and biotechnological treatments available for textile effluents will show that the latter is developing rapidly, especially because they are environmentally friendly.

2.2. Physicochemical treatments

Textile effluents can be treated in two ways; the first is to remove unmodified dyes, transferring them from the effluent to a different matrix. The second approach is to degrade the dye into a compound with lower molecular weight [25].

Coagulation/flocculation is a basic method for dye removal, in which inorganic salts are added to agglutinate particles suspended in the aqueous medium, followed by the addition of a polymer that captures the clots produced, increasing their weight to promote sedimentation [27]. Electrocoagulation consists of a series of electrolytic reactions on electrode surfaces that form clumps in the aqueous phase, which adsorb contaminants and are then removed by sedimentation [23].

Other technologies are based on the absorption of dyes in specific synthetic or natural materials, such as wood charcoal, sulfonated charcoal, powdered activated carbon, orange peel, pasteurized waste water solids, and pulverized macro-fungi [28].

With respect to dye degradation, a treatment method uses ozone to oxidize contaminants not susceptible to biodegradation [29]. Another option is the oxidation by homolytic fission of hydrogen peroxide (H_2O_2), in which the medium is irradiated with ultraviolet light to generate OH* radicals, and the latter oxidize dyes [29]. Finally, oxidation with the Fenton reagent consists of the use of H_2O_2 activated with a Fe(II) salt [25].

Despite the widespread application of physicochemical technologies, these have the disadvantage of producing high amounts of residual sludge, have high operating costs, and pose problems of secondary contamination, which limit their implementation in plants for textile effluent treatment [23].

2.3. Biotechnological approaches

Biotechnological methods for the treatment of textile waste water are based on the capacity of plants, algae, fungi, bacteria, lichens, and consortia to degrade contaminants. Microorganisms use diverse mechanisms to remove dyes, such as the aerobic or anaerobic production of enzymes (biodegradation) [8, 12] and biosorption [30]. In biosorption, heteropolysaccharides and lipids in the cell wall of some organisms are responsible for dye removal [31]. With respect to biodegradation, various microbial enzymes are known to be capable of reducing or oxidizing dyes.

Methanogenic and acidogenic bacteria have been reported to produce azo-reductases, which transform azo dyes into aromatic amines [8]. Under anaerobic conditions, the bacterial enzymes FMN-dependent reductase, FMN-free reductase, NADH-dependent reductase, NADPH-dependent reductase, and NADH-DCIP-dependent reductase are capable of degrading dyes. By contrast, the major bacterial enzymes produced under aerobic conditions are manganese peroxidase, lignin peroxidase, laccase, tyrosinase, N-demethylase, and cellobiose dehydrogenase [31].

The use of plants to remove contaminants is known as phytoremediation; dyes can be sipped, accumulated, transformed, and/or volatilized upon contact, mainly by plant roots [32]. Plants with long roots and rapid growth, such as *Aster amellus* Linn., are good candidates for the treatment of water containing azo dyes [3]. On the other hand, algae, being resistant to the conditions found in textile effluents, are also used for dye removal. Malachite green has been removed by the macroalga *Chara* sp. through degradation and sorption mechanisms [33], and by the microalga *Comarium* sp. [5]. However, a disadvantage of using plants or algae for dye removal is that both are time-consuming processes [33].

Filamentous fungi can degrade colorants thanks to their high capacity to adapt their metabolism to exploit various carbon and nitrogen sources [31]. The white rot filamentous fungus *Trametes versicolor* degrades acid red dye 27 through lignin peroxidases [34]. Other filamentous organisms such as *Aspergillus niger* and *A. terreus* degrade and adsorb the red azo dye MX-5B, reducing its toxicity [30].

Bacteria decolorize and mineralize azo compounds by combining aerobic and anaerobic processes. Azo bonds are first reduced (anaerobically) to form aromatic amines, and such amines are subsequently deaminated or dehydrogenated (aerobically) [6, 8].

Various reports on the decolorization of azo compounds with plant, fungal, and bacterial monocultures have demonstrated their limitations, as evidenced by a low enzyme production or an incomplete dye degradation of [8, 12]. A little studied biotechnological approach is to couple two microorganisms to complement their degradative capacities. A clear case of synergism is the interaction of *A. ochraceus* NCIM-1146 (fungus) with *Pseudomonas* sp. SUK1 (bacteria), which resulted in increased biodegradation and detoxification rates for the azo dye Rubine GFL [7]. In the interaction between *Glandularia pulchella* Tronc. (plant) and *P. monteilli* ANK (bacterium), 100% degradation rates for the Scarlet RR dye were attained, due to enzymatic coupling [9].

Designed consortia are "man-made" systems in which the growth of two or more organisms, chosen specifically to improve a bioprocess, is induced in the same medium [20]. There are few reports in the literature about designed consortia applied in the treatment of waste water for dye removal [14, 16, 17, 30, 35, 36].

The pioneering studies on the subject were conducted on natural consortia, obtained from sites contaminated with textile effluents. Their availability was extensive and showed higher efficiency rates than monocultures. However, due to their complexity, they are considered as "black box" systems because neither the identity of the organisms in the consortia, the interaction among them (mutualism, commensalism, amensalism, or competition) [37], nor the type of participation of each organism in the removal process are known [38]. Some of these disadvantages were evident in a fungus-bacterium biofilm that worked continuously for four months under non-sterile conditions. The results showed that *C. tropicalis* and *Candida* sp. prevailed until the end of dye removal, while all other microorganisms were suppressed or did not exhibit decolorizing activity [38].

Among the enzymes involved in the biotechnological treatment of textile effluents, laccases are the most recurrent [39]. Laccases have been reported to be overproduced in consortia, so their production could serve as an indicator of the advantages of consortia over monocultures. In this regard, the following section briefly describes the factors affecting laccase production in monocultures and consortia.

3. Monocultures and consortia in laccase production

3.1. Biological distribution

Laccase production is widespread among insects [13], plants, fungi, bacteria [40], and cyanobacteria [41]. Due to their extracellular (plants and fungi) or intracellular (bacteria) location [42], diverse physiological functions have been suggested [43]. In insects, they participate in the synthesis of epidermal cuticle. In plants, they contribute to the synthesis of lignin [44], while in fungi they participate in lignin degradation, plant pathogenesis, and competitive interactions [13]. In bacteria, they are related to pigment biosynthesis [10].

3.2. General characteristics

In general, laccases are dimeric or tetrameric glycoproteins [41], N-glycosylated glycoproteins [11] of the oxidase type (benzenediol: oxygen reductase, EC 1.10.3.2), with a molecular weight of 40-100 kDa [45, 13]. Both plant and fungal laccases are glycosylated [46]. Laccases can be classified into blue, yellow, and white laccases according to the number of copper atoms in their catalytic centers [40]. Blue laccases are the most abundant; they have four type 1 copper (T1Cu) centers, where the oxidation of reduced substrates occurs [46]. Type 2 (T2Cu) and type 3 (T3Cu) copper centers make up the trinuclear group, in which an oxygen molecule is reduced to two water molecules [47]. This way, a laccase molecule catalyzes four single-electron reduction reactions, from O_2 to H_2O, using phenolic substrates as hydrogen donors [11] and molecular oxygen as the sole co-substrate [48].

The optimal pH values and reaction temperatures for laccases vary from 2 to 10 and 40 to 65 °C, respectively [40]; this remarkable stability in wide pH and temperature ranges, due to N-glycosylation [47], is advantageous for enzyme secretion [46]. Laccase isoelectric points range between 2.6 and 6.9 [47]. With respect to redox potential (E°), which refers to the energy that enzymes require to remove an electron from the substrate, higher values of T1Cu E° indicate greater oxidation power of laccases on substrates [49]. In those cases where the value of E° is higher for the substrate than for laccases, the addition of a mediator may overcome the energetic barrier [50], extending the catalytic activity of laccases to non-phenolic substrates. While it was recently reported that there are more than 100 different laccase mediators, the most studied are ABTS (2,2'-azino-bis (3-ethyl benzothiazoline-6-sulfonic acid)) and HBT (1-hydroxybenzotriazole) [51].

3.3. Production of fungal laccases

3.3.1. Determination of laccase activity

The production of laccases is measured indirectly through laccase activity, determined by spectrophotometric techniques; a unit of laccase activity is defined as the number of micromoles of the enzyme that oxidize one micromole of substrate per minute per unit volume or mass, under standard conditions. Among the main substrates used to determine laccase activity are ABTS [52], DMP (2,6-dimethoxyphenol) [40], catechol [53], guaiacol [54], and 3,3-diaminobenzidine [14].

3.3.2. Factors that affect laccase production

Carbon and nitrogen sources control laccase production [47, 52], although to a lesser extent than chemical mediators [10], natural lignin-derived mediators [55], and biological inducers [11].

3.3.3. Laccase-producing microorganisms

Table 1 shows the factors affecting laccase production for several microorganisms, either in monocultures or consortia. Laccase production rates ranged from 57 to 8 533 000 Ul^{-1} as determined by the ABTS method, and from 1100 to 72 000 Ul^{-1} as determined with the DMP method. The fungus *Pleurotus ostreatus* (ACCC52857) has the highest laccase activity rate reported (8 533 000 Ul^{-1}) [52]; it is noteworthy that *P. ostreatus,* a wood saprophyte, produces 12 laccase isoforms [56]. Then, the yeast *Cryptococcus albidus* FIST3 was reported to produce 832 200 Ul^{-1} of laccase activity; this organism was isolated from effluents from the pulping and paper industry [39]. There are few reports about the production of laccases by *C. albidus*, probably because it is a pathogenic agent, causing encephalitis in HIV patients [57]. The filamentous fungus *Coriolopsis gallica* 1184 produced 200 900 Ul^{-1} of laccase activity [48]; this fungus was isolated from decaying plant material and has been used for the removal of phenolic compounds [58]. A production of 143 000 Ul^{-1} by the fungus *Pycnoporus sanguineus* (CS43) was reported [47]. The laccases produced by *Pycnoporus* sp. have been under study in the last decade [59] for the treatment of textile effluents [60], biotransformation of pharmaceutical microcontaminants [61], and degradation of endocrine disruptors [47].

Regarded as a natural inductor, the consortium formed by *Phelebia radiata* with *Dichomitus squalens* and *P. radiata* with *Ceriporiopsis subvemispora* (all of them white rot fungi) produced 118 000 Ul^{-1} of laccase activity. It should be noted that such high production rates were considered as a synergistic response from the consortium [62]. Monocultures of *Ganoderma lucidum* [10], *Pycnoporus* sp. SYBC-L3 [40], *Anthrospira maxima* (SAE-25780) [41], *T. versicolor* ATCC 42530 [44], *T. versicolor* CICC 14001 [53], *Xylaria* sp. [63], *Cerrena consors* [11], *T. versicolor/Candida* sp. HSD07A [64], and *Rhodotorula mucilaginosa/Pleurotus ferulae* JM301 [55] showed production rates below 100 000 Ul^{-1} of laccase activity.

In general, white rot fungi are primary laccase producers [62], probably because genetic multiplicity allows them to secrete different isoforms [13]. A review on inducers that stimulate laccase production will provide evidence that consortia are efficient biological inductors and could replace chemical inducers.

3.3.4. Inductors in laccase production

Two types of compounds have been reported to increase laccase yields: mediators and inducers. Mediators are low molecular weight molecules that are easily oxidized by laccases; they act by donating electrons to a complex molecule that laccases cannot oxidize [11]. On the other hand, inducers are complex molecules or organisms that affect the metabolism of the laccase-producing organism, stressing it [65] and impacting it at a genetic level [64].

Table 1 shows various mediators and inducers used in the production of laccases. These auxiliary mediators include $CuSO_4$ [52], vanillin [48], ethanol [10], gallic acid [10], tween 80 [64] 2,4,6-trinitrotoluene, ferulic acid, hydroquinone [66], as well as dyes such as Lanaset [67], Scarlet RR [14], and malachite green [17].

The effect of the chemical inducer paraquat was observed when it was brought into contact with *T. versicolor*. In response to oxidative stress, the fungus increased the production of antioxidant enzymes such as laccases, superoxide dismutase, and peroxidase [65]. However, the use of chemical inducers has been related to cases of toxicity; thus, a rational use of $CuSO_4$ has been suggested to prevent environmental risks [11]. Some natural mediators based on lignocellulosic materials are corn stem and ear, rice straw [62], tamarind shell [10], sawdust, and grape seeds and stems [68]. Compounds derived from lignin such as guaiacol [41], 2,5-xylidine [63], and phenolic compounds obtained from amurca [11] have also been used, even though these inducers have been reported to increase laccase production time [62].

Given the disadvantages of the methods discussed above, a biotechnologically viable approach is the use of biological inducers, which consist of fungal/fungal [17], fungal/yeast [55], and fungal/bacterial [14] consortia, among other types. Consortia have less toxic effects and allow for shorter laccase production times. The increase in laccase production when consortia are cultured has been related to: a) morphological changes and alterations in the growth patterns of the cultured organisms; b) the production of laccase isoforms; c) stress due to competition for the substrate; and c) generation of secondary metabolites. More precisely, it has been reported that when co-cultured with *R. mucilaginosa,* the fungus *P. ferula* increased the overall yield of laccases and secreted new laccase isoforms in response to a harsher competition for substrates [55]. Similarly, the consortium formed by *T. versicolor* and *Candida* sp. HSD07A showed a 1.18-fold increase in laccase production (with respect to the monoculture) because the HSD07A strain consumed 99% of the glucose in the culture medium within 10 h, leaving the fungus in starvation and ultimately increasing laccase production [64]. Additionally, there is evidence that antifungal metabolites produced by *Pseudomonas fluorescens* when co-cultured with *R. solani* caused the latter to increase laccase production [69].

Not all consortia increase laccase production, however. For instance, the consortium formed by *Pleurotus ostreatus* and *P. citrinopileatus* (both lignin degraders) decreased laccase and Mn-peroxidase production, with both organisms affecting the development of each other [70].

The first effect of consortium growth is an alteration in the patterns of enzyme production; this is beneficial in systems where there is interest in transforming enzymatically certain substances, as it is the case of dye removal. Decolorization rates are usually different in monocultures and consortia.

Preprints (www.preprints.org) | NOT PEER-REVIEWED | Posted: 20 April 2018 doi:10.20944/preprints201804.0264.v1

Table 1. Factors that affect the production of laccases

Microorganism	Carbon source	Nitrogen source	Mediator /Inductor	Laccase activity (UI^{-1})	Time (d)	Reference
Pleurotus ostreatus (ACCC52857)	Glucose	Potato extract	$CuSO_4$	8 533 000	13	[52]
Cryptococcus albidus		Meat peptone	$CuSO_4$ and bagasse	832 200	11	[39, 71]
Coriolopsis gallica 1184		Bacto peptone	Vanillin	200 900	7	[48]
Pycnoporus sanguineus (CS43)	Tomato juice		$CuSO_4$	143 000	15	[47]
Phelebia radiata/Dichomitus squalens *Phelebia radiata/Ceriporiopsis subvemispora*	Glucose and cornstarch	Peptone	Lignocellulosic compound and fungus/fungus consortium	95 000	9	[62]
	Glucose and corn			110 000	9	
	Glucose and wheat straw			88 000	12	
	Glucose and rice straw			118 000	12	
Ganoderma lucidum	Glucose	Soy extract	Tamarind shell, ethanol and $CuSO_4$, gallic acid	74 840	15	[10]
Pycnoporus sp. SYBC-L3		$NaNO_3$	$CuSO_4$	72 000	6	[40]
Anthrospira maxima (SAE-25780) (cyanobacterium)	Sucrose	$NaNO_3$	$CuSO_4$ and guaiacol	56 894	4	[41]
Trametes versicolor ATCC 42530	Glucose	NH_4Cl	Without induction	50 660 ($Ul^{-1}h^{-1}$)	100 h	[44]
Trametes versicolor CICC 14001			Ultrasound waves	23 140	3	[53]
Xylaria sp.	Wheat bran	$(NH_4)_2SO_4$	2,5-xylidine	20 535	16	[63]
Cerrena consors	Amurca	Malt extract agar	Amurca	1350	30	[11]
			$CuSO_4$ + Amurca	13 055	25	
Cerrena consors/Bionectria ochroleuca			Fungus/fungus consortia	2831	25	
Cerrena consors/Lasiodiplodia theobromae				2865	25	
Trametes versicolor/Candida sp. HSD07A	Glucose	$(NH_4)_2C_4H_4O_6$	Tween 80 and fungus/fungus consortia	10 500	6	[64]
Rhodotorula mucilaginosa/Pleurotus ferulae JM301	Glucose, wheat bran, and corn flour	Wheat bran and maize flour	Lignocellulose compounds and fungus/yeast consortium	10 055	8	[55]
Trametes sp. AH28-2/*Trichoderma* sp. ZH1	Xylose	Tryptone	Fungus/fungus consortia	6210	8	[54]
Trametes versicolor ATCC 42530	Glucose	NH_4Cl	Lanaset G	1700	4	[73]
				2000	20	
Trametes trogii LK13	Rice straw, bagasse, sawdust, and fragments of cotton seed coat	Peptone and malt extract	Lignocellulosic material and $CuSO_4$	1263 Ug^{-1}	7	[76]
Trametes versicolor ATCC 42530	Glucose	NH_4Cl	Lanaset G	1100	6	[67]
Trametes versicolor HEMIM-9	White wheat flour and cereal flakes		Sawdust	800	48 h	[45]
Trametes versicolor (CBS100.29)	Glucose	Lignocellulosic material	Grape seeds	250	35	[68]
			Grape stems	400		
			Barley bran	650		
Trametes versicolor CICC 14001		NH_4Cl	Ultrasound waves	588.9	5	[53]

Streptomyces cyaneus	Soy flour	$(NH_4)_2SO_4$	$CuSO_4$	57	20	[77]
Trametes versicolor BAFC 42FC/*Ganoderma lucidum* E47	Oat seeds		Lignocellulosic material and fungus/fungus consortia	7.93 Ug^{-1}	14	[17]
Trametes versicolor G3 (DMS 11269)	Glucose	$(NH_4)_2C_4H_4O_6$	Lignocellulosic compounds	0.3 Ug^{-1} biomass	12	[78]
	Wheat straw			14 Ug^{-1} biomass	7	
	Wood chips			13.5 Ug^{-1} biomass	6	
Galactomyces geotrichum MTCC 1360/*Brevibacillus laterosporus* MTCC 2298	Malt extract and nutritious broth		Colorants and fungus/bacteria consortium	0.372 Umg^{-1} of protein	18 h	[14]

3.4. Monocultures and consortia in dye decolorization/degradation

3.4.1. Difference between decolorization and biodegradation

Decolorization is defined as the elimination or transformation of the chromophore group in a compound [30]. Biodegradation, on the other hand, consists in decomposing the dye by biological means, while reducing its molecular weight and the complexity of its chemical structure [36].

3.5. Dye decolorization/biodegradation

Different approaches have been reported to decolorize and biodegrade dyes; among them are: 1) enzymatic extracts, 2) enzymes immobilized in polymer matrices, 3) enzyme-mediator systems (either free or immobilized), and 4) growing monocultures and consortia (Table 2).

In a comparison between the enzymatic decolorization of 150 mg l^{-1} of Gris Lanaset G (GLG) in media either seeded or not with *T. versicolor*, decolorizing rates of 90% and 35%, respectively, were obtained. Fungal cells could metabolize dye derivatives to increase enzymatic production [67]. Both the microorganism and decolorizing enzymes exhibited poor stability under the adverse conditions of the medium (textile effluent), so encapsulating or immobilizing them in suitable materials improved their stability. Waste water from a cellulose plant was treated with *T. versicolor* cells, both free and immobilized in nylon [44]. Immobilized cells were more efficient to treat waste water, reducing color (36%), concentration of aromatic compounds (54%), and toxicity. In another study, laccases from *T. versicolor* were immobilized in mesoporous walnut shell charcoal to scale up dye treatment; this method was able to decolorize both acid and reactive dyes [47].

Laccase-mediator systems are efficient in the decolorization of azo dyes. The dye red acid 97 was decolorized by 90% in three minutes by 500 Ul^{-1} of laccase from *T. histuria* (BT 2566) in the presence of violuric acid, while decolorization rate was 30% in 1.5 h in the absence of the mediator [75]. Laccase activity is increased by mediators due to the presence of electron-donor substituents in the benzene ring, which reduce E° [79]. Mediator-laccase systems have been used for waste water treatment in the paper pulping and bleaching industry [80], in the bioremediation of PHAs, azo dyes [50], and sulfonamide antibiotics [81], among others. While inexpensive mediators are available, such as N-hydroxyacetanilide (HNA) [80], their high toxicity and our lack of knowledge on their effects limit the use of laccase chemical-mediator systems.

3.6. Decolorization/biodegradation with consortia

Few practical applications of fungal or bacterial monocultures for the removal of azo dyes have been reported. Their disadvantages are due to their susceptibility to biological contamination [15], low production of decolorizing enzymes [17], poor adaptation to the complex and variable abiotic conditions of textile effluents [18], enzymatic destabilization [19], the fact that few fungi use dyes as a carbon and energy source [82], and low efficiency rates for bacterial degradation in aerobic conditions [19]. Microbial consortia show clear advantages over monocultures for dye removal [16, 14], as will be discussed below.

Some studies on dye treatment with consortia are shown in Table 2. The increase in decolorization efficiency can be exemplified in the study of the consortium formed by *Galactomyces geotrichum* MTCC 1360 and *Bacillus* sp. VUS, for which a 100% decolorization of the azo compound Brown 3REL was obtained in 2 h; in contrast, significantly lower decolorization rates (39% in 24 h) and higher staining time (100% in 5 h) were obtained with *G. geotrichum* MTCC 1360 and *Bacillus* sp. VUS monocultures, respectively [16]. Similarly, a more significantly higher malachite green decolorization rate has been reported by the consortium formed by *G. lacidum* and *T. versicolor* with respect to monocultures, as a result of the secretion of new laccase isoforms [17]. A new metabolic pathway was determined to degrade the dye Scarlet RR by the consortium formed by *G. geotrichum* MTCC 1360 and *Brevibacillus laterosporus* MTCC 2298; changes in the production levels of versatile alcohol oxidase, laccase, tyrosinase, and NADH-DCIP reductase were observed with respect to monocultures [14].

Usually, the role of each member of designed consortia in dye transformation is known. This is the case of the bacterial consortium NAR-2, formed by *Citrobacter freundii* A1, *Enterococcus casseliflavus* C1, and *Enterobacter cloacae* L17, which degraded the dye acid red 27. The fate of substrates and intermediate metabolites was explored, and it was demonstrated that the acid red 27 undergoes an amination and desulfonation process during a stage of microaerophilic biodegradation, followed by an azo reduction (the actual decolorization step, lasting 2 h). Subsequently, a mineralization phase took place under aerobic conditions (lasting 48 h) [36].

Additionally, the capacity of the organisms to produce decolorizing enzymes under aerobic or anaerobic conditions is changed when the consortium is formed [82]. *Pseudomonas* sp. SUK1 decolorizes the red compound BLI [83] and has been reported to decolorize the dye RNB HE2R in consortium with *Aspergillus ochraceus* NCIM-1146, but under anaerobic conditions only [84]. However, in consortium with *A. ochraceus* NCIM-1146, *Pseudomonas* sp. SUK1 was able to degrade the dye Rubine GFL from a textile effluent in an aerated liquid system [7].

Table 2. Studies on decolorization and biodegradation of dye-containing effluents by monocultures and consortia.

Microorganisms	Dyes in effluent	Treatment conditions	Results	Decolorizing agent	Reference
Cryptococcus albidus	1.0% textile effluent and 0.1% dyes: aniline blue, xylene cyanol, bromothymol blue, carmine, crystal violet, Coomassie brilliant blue R-250, and trypan blue. At 0.1%: tetrachlorohydroquinone, 4-chlorosalisilic acid, 3-methyl-catechol, 2,4-dichlorophenol, and hydroquinone.	Sodium tartrate (85 mM, pH = 3), plus 2 Ul^{-1} of laccase activity at 3 °C for 1 h.	Carmine, crystal violet, and aniline blue removal by 40%; all other compounds by < 30%.	Laccase extract	[72]
Ganoderma lucidum	Methyl violet 2B (MV), Remazol Yellow G (RY) and Acid-fast red (AFR) 50 mM	20 Uml^{-1} of laccase activity in sodium acetate 100 mM (pH = 5) for 24 h.	MV removal by 78%, RY by 83%, AFR by 92%.	Laccase extract	[10]
	Effluent from dye industry	Culture medium (pH = 5.5) for 21 days.	97% decolorization, DBO and DQO removal by 75% and 70%, respectively.	Fungal growth-produced laccases	
Trametes versicolor (CBS100.29)	Phenol red (75 μM)	72 Ul^{-1} of laccase activity with phenol red plus sodium acetate 10 mM (pH = 4.5) at 30 °C.	61% decolorization in 72 h.	Laccase extract	[68]
Trametes versicolor ATCC 42530	Effluent from cellulose plant	Culture medium with effluent (pH = 4.5), 2,2-dimethyl succinate and inoculum immobilized in polyurethane foam.	Color reduction by 36%, aromatic compounds by 54%, and 3.15-fold reduction in toxicity.	Immobilized microorganism and fungal growth-produced decolorizing enzymes	[44]
Trametes versicolor ATCC 42530	Gris Lanaset (GLG)	100 ml of laccase solution (2500 Ul^{-1}) at 25 °C, 135 rpm, 150 mg l^{-1} of GLG (pH = 4.5).	Decolorization by 90%.	Laccase extract	[67]
	Synthetic effluent with GLG	Sterile synthetic waste water inoculated with 3.2 gl^{-1} dry weight fungus.		Growth-produced fungus and laccases	[73]
Trametes versicolor	Acid orange 7 (AO7), Acid blue 74 (AB74), Reactive red 2 (RR2), and Reactive black 5 (RB5)	Lots at 150 rpm, 25 °C, 3 mg ml^{-1} of enzyme dissolved in phosphate buffer (100 mM, pH 6), 200 mg l^{-1} of dye.	AO7 and AB74 decolorized by 90%.	Purified laccases	[47]
Trametes versicolor CNPR 8107	Remazol blue RR and Remazol red RR	Dye (1.8 gl^{-1}) in Kirk medium incubated for six days at 30 °C.	Remazol blue RR and Remazol red RR decolorized by 96%.	Growth-produced fungal biomass, laccases and Mn-peroxidases.	[74]
Trametes versicolor ATCC 20869	Acid red 27	Culture medium added with 1 gl^{-1} of acid red 27, incubated for 4 d.	100% decolorization.	Growth-produced fungal biomass, laccases and Mn-peroxidases.	[34]

Preprints (www.preprints.org) | NOT PEER-REVIEWED | Posted: 20 April 2018 doi:10.20944/preprints201804.0264.v1

Trametes hirsuta (BT 2566)	9.2% mixture of acid red 97 (AR 97), acid green 26 (AG 26) and copper phthalocyanine.	Violuric acid added with 500 Ul^{-1} of laccase activity in phosphate buffer; pH = 5 for AR 97 (40 mgl^{-1}); pH = 4 for AG 26 (130 mgl^{-1}). Room temperature, no incubation.	AR 97 decolorized by 90% at 3 min with violuric acid 2 mM, and for AG 26 by 6.2% at 24 h.	Laccase-mediator system	[75]
Aspergillus niger *Aspergillus terreus*	Procion red MX-5B	Stage 1, biosorption: dye 200 μg ml^{-1}, pH 4, 3 mg ml^{-1} biomass Stage 2, biodegradation: dye 200 μg ml^{-1}, pH 4, 3 mg ml^{-1} biomass.	Removal of chromophore groups and decreased toxicity. Biodegradation by 98% at 336 h.	Fungal biomass	[30]
Aspergillus lentulus, *Aspergillus terreus*, and *Rhizopus oryzae*	Cu^{2+}, Cr^{6+}, acid blue 161 (AB), and Pigment orange 34 (PO)	Culture medium added with 100 mgl^{-1} of metals or dyes plus spore suspension 1% at 30 °C, 150 rpm, for 48 h.	Cr^{6+} removal by 100%, Cu^{2+} by 81.6%,AB by 98% and PO by 100%.	Consortium growth-produced microorganisms and enzymes	[35]
Trametes versicolor BAFC42FC/*Ganoderma Lucidum*	Malachite green	50 μM dye in acetate buffer (pH = 3.6)	Removal by 80% in 3 h.	Growth-produced microorganisms and enzymes	[17]
Galactomyces geotrichum MTCC 1360/*Brevibacillus laterosporus* MTCC 2298	Sulfonic, azoic, reactive, and dispersed dyes	20 ml of pre-grown *B. laterosporus* culture and 2 g of *G. geotrichum* biomass, 80 ml of effluent.	BOD and COD removal by 68% and 74%, respectively, in 48 h.	Consortium growth-produced biomass and enzymes	[14]
Galactomyces geotrichum MTCC 1360/*Brevibacillus laterosporus* MTCC 2298	Scarlet RR	Consortium culture added with 50 mg l^{-1} of dye	Color reduction by 98% in 16 h.	Increased production of veratril alcohol oxidase, tyrosinase, laccase, and NADH-DCIP	[14]
Citrobacter freundii A1, *Enterococcus casseliflavus* C1, and *Enterobacter cloacae* L17 (bacteria/bacteria/bacteria)	Acid red 27	Consortium culture added with 0.1 gl^{-1} of dye at 45 °C to decolorize and 37 °C, 200 rpm, to degrade.	100% removal: decolorization/degradation: 20 min/48h.	Consortium microorganisms	[36]
Galactomyces geotrichum MTCC 1360/*Bacillus* sp. VUS	Brown 3REL, Brilliant blue G, Navy blue, Yellow brown and Remazol red, 50 mg l^{-1} each.	Static cultures at 50 ° C, pH = 7.	100% decolorization in 24, 9 and 8 h for Brilliant Blue G, Navy blue, and Brown 3REL, respectively.	Consortium growth-produced enzymes	[16]
	Brown 3REL	Cultures at 150 rpm, 50 °C, pH = 7.	100% decolorization in 2 d.	Lignin peroxidase, tyrosinase, and riboflavin reductase	

As shown above, microorganisms can adapt their metabolism to meet the consortium main objective. In the designed consortium formed by *Escherichia coli* DH5*α* and *Pseudomonas luteola,* the decolorization of Reactive red 22 was due to enzymes produced by *P. luteola*, while the role of *E. coli* DH5*α* was to release extracellular metabolites that acted as mediators. Since the organisms are under stress in the presence of contaminants, both produced secondary metabolites to detoxify their environment and promote their survival [82].

Often, the products of dye degradation are toxic and require to be transformed into harmless compounds [85]. It should be noted that after degradation of acid red 27 by *T. versicolor*, FTIR analysis showed the disappearance of azo group signals, while peaks related to compounds such as naphthalene and substituted benzene rings appeared [34]. The aromatic amines resulting from the decomposition of sulfonated azo dyes are more difficult to degrade due to the hydrophilic nature of the sulfonate groups, which hinder their transport through the cell membrane [86]. Reports in the literature on consortia designed for the removal of decolorization derivatives are scarce. On the other hand, some reports described the degradation of aromatic compounds by monocultures. For example, the halophilic anaerobic eubacteria *Haloanaerobium prevalen*t DMS 2228 and *Sporohalobacter marismortui* ATCC 35420 degrade nitro-substituted aromatic compounds to the corresponding amines, such as nitrobenzene, o-nitrophenol, m-nitrophenol, p-nitrophenol, nitroanilines, 2, 4-dinitrophenol, and 2,4-dinitroaniline [87]. Analogously, *Pseudomonas putida* B2 degrades o-nitrophenol and m-nitrophenol with the subsequent release of nitrite and ammonium, respectively. The *P. putida* B2 strain of employs an oxidative pathway to degrade o-nitrophenol and a reductive pathway for m-nitrophenol [88]. Accordingly, the inclusion of strains producing decolorizing enzymes and bacteria capable to degrade aromatic compounds should be considered in the design of new consortia.

4. Conclusions

Due to their limitations, chemical and natural mediators are being replaced by consortium cultures for laccase production. Consortia have proved to induce laccase production and the secretion of new laccase isoforms. This increase in laccase yields in consortium cultures could be related to morphological changes and alterations in the growth patterns of consortium members, a competition for the substrate, and the generation of secondary metabolites that stimulate the growth of producer fungi. Consortia improve dye removal from waste water by producing enzymes that act synergistically and allow a metabolic adaptation that changes the patterns of enzymatic production, promoting different ways of dye degradation. A new approach in consortia design for waste water treatment should consider to include fungi as producers of decolorizing enzymes and bacteria as degraders of decolorization-derived compounds. Thus, consortium design provides new tools and generates new technological approaches for the remediation of textile effluents.

Author Contributions: All authors were involved in data analysis and the preprocessing phase, simulation, result analysis and discussion, and manuscript preparation. All authors approved the submitted manuscript. All authors equally contributed to writing the paper.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Cattoor, T. European legislation relating to textile dyeing. In T. Institute, *In Environmental aspects of textile dyeing*. Centexbel Belgium: Christie R. M. **2007**, 1-29.
2. Boyter, H. A. Environmental legislation USA. In I. o. Textile, In *Environmental aspects of textile dyeing* edited by the USA: Christie R. M. **2007**, 30-43.
3. Khandare, R. V.; Kabra, A. N.; Tamboli, D. P.; Govindwar, S. P. The role of *Aster amellus Linn*. in the degradation of sulfonated azo dye Remazol Red: A phytoremediation strategy. *Chemosphere*. **2011**, 82, 1147-1154.

Preprints (www.preprints.org) | NOT PEER-REVIEWED | Posted: 20 April 2018 doi:10.20944/preprints201804.0264.v1

4. Kulkarni, A. N.; Kadam, A. A.; Kachole, M. S.; Govindwar, S. P. *Lichen perlata*: A novel system for biodegradation and detoxification of disperse dye Solvent Red 24. *J. Hazard. Mater.* **2014**, 276, 461-468.
5. Daneshvar, N.; Ayazloo, M.; Khataee, A. R.; Pourhassan, M. Biological decolorizarion of dye solution containing Malachite Green by microalgae *Cosmarium sp*. *Bioresour. Technol.* **2007**, 98, 1176-1182.
6. Garg, S. K.; Tripathi, M.; Singh, S. K.; Tiwari, J.Bio Decolorization of textile dye effluent by *Pseudomonas putida* SKG-1 (MTCC 10510) under the conditions optimized for monoazo dye orange II color removal in a simulated minimal salt medium. *Int. Biodeterior.Biodegrad.* **2012**, 74, 24-35.
7. Lade, H. S.; Waghmode, T. R.; Kadam, A. A.; Govindwar, S. P. Enhanced biodegradation and detoxification of disperse azo dye Rubine GFL and textile industry by a defined fungal-bacterial consortium. *Int. Biodeterior.Biodegrad.* **2012**, 72, 94-107.
8. Pandey, A.; Singh, P.; Iyengar, L. Bacterial decolorization and degradation of azo dyes. *Int. Biodeterior.Biodegrad.* **2007**, 59, 73-84.
9. Kabra, A. N.; Khandare, R. V.; Govindwar, S. P. Development of a bioreactor for remediation of textile effluent and dye mixture: A plant-bacterial synergistic strategy. *Water Res.* **2013**, 47, 1035-1048.
10. Manavalan, T.; Manavalan, A.; Thangavelu, K. P.; Heese, K. Characterization of optimized production, purification, and application of laccase from *Ganoderma Lucidum*. *Biochem. Eng. J.* 2013, 70, 106-114.
11. Mann, J.; Markham, J. L.; Peiris, P.; Spooner-Hart, R. N.; Holford, P.; Nair, N. G. Use of olive mill wastewater as a suitable substrate for the production of laccase by *Cerrena consors*. *Int. Biodeterior.Biodegrad.* **2015**, 99, 138-145.
12. Baldrian, P.An increase of laccase activity during an interspecific interaction of white-rot fungi. *FEMS Microbiol. Ecol.* **2004**, 50, 245-253.
13. Rivera-Hoyos, C. M.; Morales-Álvarez, E. D.; Poutou-Pinales, R. A.; Pedroza-Rodríguez, A. M.; Rodríguez-Vázquez, R.; Delgado-Boada, J. M. Fungal laccases. *Fungal Biol. Rev.* **2013**, 27, 67-82.
14. Kurade, M. B.; Waghmode, T. R.; Kagalkar, A. N.; Govindwar, S. P. Decoloration of textile industry effluent containing disperse dye Scarlet RR by a newly developed bacterial-yeast consortium BL-GG. *Chem. Eng. J.* **2012**, 184, 33-41.
15. Kurade, M. B.; Waghmode, T. R.; Kagalkar, A. N.; Govindwar, S. P. Decoloration of textile industry effluent containing disperse dye Scarlet RR by a newly developed bacterial-yeast consortium BL-GG. *Chem. Eng. J.* **2012**, 184, 33-41.
16. Jadhav, S. U.; Jadhav, U. U.; Dawkar, V. V.; Govindwar, S. P. Biodegradation of Disperse Dye Brown 3REL by Microbial Consortium of *Galactomyces geotrichum* MTCC 1360 and Bacillus sp. VUS. *Biotech.Bioprocess Eng.* ***2008***, 13, 232-239.
17. Kuhar, F.; Castiglia, V.; Levin, L.Enhancement of laccase production and malachite green decolorization by co-culturing *Ganoderma Lucidum* and *Trametes versicolor* in solid-state fermentation. *Int. Biodeterior. Biodegrad.* **2015**, 104, 238-243.
18. O´Neill, C.; Hawkes, F. R.; Hawkes, D. L.; Lourenco, N. D.; Pinheiro, H. M.; Delée, W. Colour in textile effluents-sources, measurements, discharge consents, and simulation: a review. *J. Chem. Technol. Biotechno.* **1999**, 74, 1009-1018.
19. Ayed, L.; Khelifi, E.; Ben, J. H.; Miladi, H.; Cheref, A.; Achour, S. Response surface methodology for decolorization of azo Methyl Orange by the bacterial consortium: Produced enzymes and metabolites characterization. *Chem. Eng.J.* **2010**, 165, 200-208.
20. Nadell, C. D.; Foster, K. R.; Xavier, J. B. The emergence of spatial structure in cell groups and the evolution of cooperation. *PLOS Computational Biology*, **2010**, 6(3): e1000716. doi:10.1371/journal.pcbi.1000716.
21. Robinson, T.; McMullan, G.; Marchant, R.; Nigam, P. Remediation of dyes in textile effluent: a critical review of current treatment technologies with a proposed alternative. *Bioresour. Techn.* **2001**, 77, 247-255.
22. Savin, I. I.; Butnaru, R. Wastewater characteristics in textile finishing mills. *Environ. Eng.Manag. J.*, **2008**, 6, 278-285.
23. Bayramoglu, M.; Kobya, M.; Can, O. T.; Sozbir, M. Operating cost of electrocoagulation of textile dye wastewater. *Sep. Purif. Technol.* **2004**, 37, 117-125.
24. Mansour, H. B.; Corroler, D.; Barillier, D.; Ghedia, K. Evaluation of genotoxicity and pro-oxidant effect of azo dyes: Acids yellow 17, violet y and orange 52, and of their degradation products by *Pseudomonas putida* mt-2. *Food Chem. Toxico.* **2007**, 45, 1670-1677.
25. Slokar, S. P.; Marechal, M. L. Methods of Decoloration of Textile Wastewaters. *Dyes Pigments.* **1998**, 37 (4), 335-356.

26. Wang, Y. Q.; Zhang, H. M.; Tang, B. P. The interaction of C.I. acid red 27 with human hemoglobin in solution. *J. Photochem. Photobiol., B.* **2010**, 100, 76-83.
27. Bidhendi, G. R.; Torabian, A.; Ehsani, H.; Razmkhah, N. Evaluation of industrial dyeing wastewater treatment with coagulants and polyelectrolyte as a coagulant aid. *Iran. J. Environ. Health. Sci. Eng.* **2006**, 4(1), 29-36.
28. Forgacs, E.; Cserháti, T.; Oras, G. Removal of synthetic dyes from wastewaters: a review. *Environ. Int.* **2004**, 30, 953-971.
29. Arslan-Alaton, I.; Alaton, I. Degradation of xenobiotics originating from the textile preparation, dyeing, and finishing industry using ozonation and advanced oxidation. *Ecotoxicol. Environ. Saf.* **2006**, 68, 98-107.
30. Almeida, E. J.; Corso, C. R. Comparative study of toxicity of azo dye Procion Red MX-5B following biosorption and biodegradation treatments with the fungi *Aspergillus niger* and *Aspergillus terreus*. *Chemosphere.* **2014**, 112, 317-322.
31. Solís, M.; Solís, A.; Inés, P. H.; Manjarrez, N.; Flores, M. Microbial decolourization of azo dyes: A review. *Process Biochem.* **2012**, 47, 1723-1748.
32. Delgadillo-López, A. E.; González-Ramírez, C. A.; Prieto-García, F.; Villagómez-Ibarra, J. R.; Acevedo-Saldoval, O. Phytoremediation: an alternative to eliminate pollution.*Trop. Subtrop. Agroecosyst.* **2011**, 14, 597-612.
33. Khataee, A. R.; Dehghan, G.; Ebadi, A.; Zarei, M.; Pourhassan, M. Biological treatment of a dye solution by Macroalgae *Chara sp.*: Effect of operational parameters, intermediates identification and artificial neural network modeling. *Bioresour.Technol.* **2010**, 2252-2258.
34. Gavril, M.; Hodson, P. V. Chemical evidence for mechanism of biodecoloration of Amaranth by *Trametes versicolor*. *World J. Microbiol. Biotechnol.* **2007**, 23, 103-124.
35. *Gavril, M. y Hodson, P. V. (2007). Chemical evidence for mechanism of biodecoloration of Amaranth by Trametes versicolor. World J Microbiol Biotechnol 23, 103-124.*
36. Chan, G. F.; Rashid, N. A.; Chua, L. S.; Ab.llah, N.; Nasiri, R.; Ikubar, M. R. Communal microaerophilic-aerobic biodegradation of amaranth by novel NAR-2 bacterial consortium. *Bioresour. Technol.* **2012**, 105, 48-59.
37. Ghrosh, S.; Chowdhury, R.; Bhattacharya, P. Mixed consortia in bioprocesses: role of microbial interactions. *Appl. Microbiol. Biotechnol*. **2016**, DOI 10.1007/s00253-016-7448-1.
38. Cui, D.; Li, G.; Zhao, D.; Gu, X.; Wang, C.; Zhao, M. Microbial community structures in mixed bacterial consortia for azo dye treatment under aerobic and anaerobic conditions. *J. Hazard. Mater.* **2012** , (221-222), 185-192.
39. Singh, R. L.; Singh, P. K.; Singh, R. P. Enzymatic decolorization and dedradation of azo dyes- A review. *Int. Biodeterior. Biodegrad.* **2015**, 104, 21-31.
40. Liu, J.; Cai, Y.; Lioa, X.; Huang, Q.; Hao, Z.; Hu, M. Efficiency of laccase production in 65-L air-lift reactor for potential green industrial and environmental application. *J. Clean Prod.* **2013**, 39, 154-160.
41. Afreen, S.; Anwer, R.; Singh, R.K.; Fatma, T. Extracellular laccase production and its optimization from *Arthrospira maxima* catalyzed decolorization of synthetic dyes. *Saudi J. Biol. Sci.* **2016**, http://dx.doi.org/10.1016/j.sjbs.2016.01.015
42. Diamantidis, G.; Effosse, A.; Potier, P.; Bally, R. Purification and characterization of the first bacterial laccase in the rhizospheric bacterium *Azospirillum lipoferum*. *Soil Biol. Biochem.* **2000**, 32, 919-927.
43. Asgher, M.; Bhatti, H. N.; Ashraf, M.; Legge, R. L. Recent developments in biodegradation of industrial pollutants by white rot fungi and their enzyme system. *Biodegradation,* **2008**, 19, 771-783.
44. Font, X.; Caminal, G.; Gabarreli, X.; Vicent, T. Treatment of toxic industrial wastewater in fluidized and fixed-bed batch reactors with *Trametes versicolor*: influence of immobilisation. *Environ. Technol.* **2006**, 845-854.
45. Bertrand, B.; Martínez-Morales, F.; Tinoco-Valencia, R.; Rojas, S.; Acosta-Urdapilleta, L.; Trejo-Hernández, M. R. Biochemical and molecular characterization of laccase isoforms produced by the white-rot fungus *Trametes versicolor* under submerged culture conditions. *J. Mol. Catal. B: Enzym.* **2015,** 122, 339-347.
46. Dwivedi, U. N.; Singh, P.; Pandey, V. P.; Kumar, A. Structire-function relationship among bacterial, fungal and plant laccases. *J. Mol. Catal. B: Enzym.* **2011**, 117-128.
47. Ramírez-Cavazos, L. I.; Junghanns, C.; Nair, R.; Cárdenas-Chávez, D. L.; Hernández-Luna, C.; Agathos, S. N. Enhanced production of thermostable laccase from a native strain of *Pycnoporus sanguineus* using central composite design. *J Zhejiang Univ-Sci B (Biomed and Biotechnol).* **2014**, 15 (4), 343-352.

48. Songulashuili, G.; Flahaut, S.; Demarez, M.; Tricot, C.; Bauvoris, C.; Debaste, F. High yield production in seven days of *Coriolopsis gallica* 1184 laccase at 50 L scale; enzyme purification and molecular characterization. *Fungal Biol.* **2016**, 120, 481-488.
49. Frasconi, M.; Favero, G.; Boer, H.; Koivula, A.; Mazzei, F. Kinetic and biochemical properties of high and low redox potential laccases from fungal and plant origin. *Biochim. Biophys. Acta* 1804, **2010**, 899-908.
50. Baldrian, P. Fungal laccases-occurrence and properties. *FEMS Microbiol. Rev.* **2005**, 30, 215-242.
51. Bourbonnais, R.; Paice, M. G. Oxidation of non-phenolic substrates. *FEMS* **1990**, 267 (1), 99-102.
52. Zhu, C.; Bao, G.; Huang, S. Optimization of laccase production in the white-rot fungus *Pleurotus ostreatus* (ACCC52857) induced through yeast extract and copper. *Biotechnol. Biotechnol. Equip.* **2016**, 30 (2), 270-276.
53. Wang, F.; Chen, G.; Liu, C.-Z. Inmobilization of *Trametes versicolor* cultures for improving laccase production in bubble column reactor intensified by sonication. *J. Ind. Microbiol. Biotechnol.* **2013**, 40, 141-150.
54. Zhang, H.; Hong, Y. Z.; Xiao, Y. Z.; Yuan, J.; Tu, X. M.; Zhang, X. Q. Efficient production of laccases by *Trametes sp.* AH28-2 in cocultivation with a *Trichoderma* strain. *Appl. Microbial. Biotechnol*. **2006**, (73), 89-94.
55. Wang, H.; Peng, L.; Ding, Z.; Wu, J.; Shi, G. Stimulated laccase production of *Pleurotus feulae* JM301 fungus by *Rhodotorula mucilaginosa* yest in co-culture. *Process Biochem.* **2015**, (50), 901-905.
56. Castanera, R.; Pérez, G.; Omarini, A.; Alfaro, M.; Pisabarro, A. G.; Faraco, V. Transciptional and enzymatic profiling of *Pleuratus ostreatus* laccase gene in sumerged and solid-state fermentation cultures. *Appl Environ. Microbiol.* **2012**, 78 (11), 4037-4045.
57. Liu, Y.; Ma, S.; Wang, X.; Xu, W.; Tang, J. *Cryptococcus albidus* encephalitis in newly diagnosed HIV-patient and literature review. *Med. Mycol. Case Rep.* **2014**, 3, 8-10.
58. Daâssi, D.; Belbahri, L.; Vallat, A.; Woodward, S.; Nasri, M.; Mechichi, T.Enhanced reduction of phenol content and toxicity in olive mill wastewater by newly isolated strain of *Coriolopsis gallica*. *Environ. Sci. Pollut. Res.* **2014**, 21, 1746-1758.
59. Lomascolo, A.; Uzan-Boukhris, E.; Herpoel-Gimbert, I.; Sigoillot, J.-C.; Lesage-Meessen, L. Peculiarities of *Pycnoporus* especies for applications in biotechnology. *Appl. Microbiol. Biotechnol.* **2011**, 92, 1129-1149.
60. Trovaslet, M.; Enaud, E.; Guiavarc'h, Y.; Corbisier, A.-M.; Vanhulle, S. Potential of a *Pycnoporus sanguineus* laccase in bioremediation of a wastewater and kinetic antivation in the presence of an anthraquinonic acid dye. *Enzyme Microb. Technol.* **2007**, 41, 368-376.
61. Rodríguez-Delgado, M.; Orona-Navar, C.; García-Morales, R.; Hernandez-Luna, C.; Parra, R.; Manlknecht, J. Biotrasformation kinetics of pharmaceutical and industrial micropollutants in groundwater by laccase cocktail from *Pycnoporus sanguineus* CS43 fungi. *Int. Biodeter.Biodegrad.* **2016**, 108, 34-41.
62. Dong, Y.-C.; Wang, W.; Hu, Z.-C.; Fu, M.-L.; Chen, Q.-H. The synergistic effect on production of lignin-modifying enzymes through sumerged co-cultivation of *Phlebia radiata*, *Dichomitus squalens* and *Ceriporiopsis subvermispora* using agricultural residues. *Bioprocess Biosyst. Eng.* **2012**, 35, 751-760.
63. Castaño, J. D.; Cruz, C.; Torres, E. Optimization of the production, purification and characterization of a laccase from the native fungus *Xylaria sp*. *Biocatal. Agric. Biotechnol.* **2015**, 4, 710-716.
64. Hailei, W.; Guangli, Y.; Ping, L., Yanchang, G.; Jun, L.; Guosheng, L. Overproduction of *Trametes versicolor* laccase by making glucose starvation using yeast. *Enzyme Microb. Technol.* **2009**, 45, 146-149.
65. Jaszek, M.; Grzywnowicz, K.; Malarczyk, E.; Leonowicz, A. Ehanced extracellular laccase activity as a part of the response system of white rot fungi: *Trametes versicolor* and *Abortiporus biennis* to paraquat-caused oxidative stress condition. *Pestic. Biochem. Physiol.* **2006**, 85, 147-154.
66. Elisashvili, V.; Kachlishvili, E.; Khardziani, T.; Agathos, S. N. Effect of aromatic compounds on the production of laccase and manganese peroxidase by white-rot basidiomycetes. *J. Ind. Microbiol. Biotechnol.* **2010**, 37, 1091-1096.
67. Romero, S.; Blánquez, P.; Caminal, G.; Font, X.; Sarrá, M.; Gabarrell, X. Different approaches to improving the textile dye degradation capacity of *Trametes versicolor*. *Biochem. Eng. J.* **2006**, 31, 42-47.
68. Lorenzo, M.; Moldes, D.; Couto, S. R.; Sanromán, A. Improving laccase production by employing different lignocellulosic waste in sumerged cultures of *Trametes versicolor*. *Bioresour. Technol.* **2002**, 82, 109-113.
69. Crowe, J. D.; Olsson, S. Induction of laccase activity in *Rhizoctonia solani* by Antagonistic *Pseudomonas fluorescens* Strains and a Range of Chemical Treatments. *Am. Soc. Microbiol.* **2001**, 67(5), 2088-2094.
70. Carabajal, M.; Levin, L.; Albertó, E.; Lechner, B. Effect of co-cultivation of two *Pleurotus* species on lignocellulolytic enzyme production and muschroom fructification. *Int. Biodeter.Biodegrad.* **2012**, 66, 71-76.

71. Singhal, A.; Choudhary, G.; Thakur, I. S. Characterization of laccase activity produced by *Cryptococcus albidus*. *Prep. Biochem. Biotechnol.* **2012**, 42, 113-124.
72. Singhal, A.; Choudhary, G.; Thakur, I. S. Optimization of growth media for enhanced production of laccase by *Cryptococcus albidus* and its application for bioremediation of chemicals. *Can. J. Civ. Eng.* **2009**, 36, 1253-1264.
73. Blánquez, P.; Casas, N.; Font, X.; Gabarrell, X.; Sarrá, M.; Caminal, G. Mechanism of textile metal dye biotrasformation by *Trametes versicolor*. *Water Res.* **2004**, 38, 2166-2172.
74. Toh, Y.-C.; Yen, J.; Obbard, J. P.; Ting, Y.-P. Decoloration of azo dyes by white rot fungi (WRF) isolated in Singapore. *Enzym. Microb. Technol.* **2003**, (30), 569-575.
75. Couto, S. R.; Sanromán, M. Á. The effect of violuric acid on the decolourization of recalcitrant dyes by laccase from *Trametes hirsuta*. *Dyes Pigm.* **2007**, 74.
76. Yan, J.; Chen, Y.; Niu, J.; Chen, D.; Chagan, I. Laccase produced by thermotolerant strain of *Trametes trogii* LK13. *Braz. J. Microbiol.* **2015**, 46 (1), 59-65.
77. Margot, J.; Bennati-Granier, C.; Maillard, J. P., B.; Barry, D. A.; Holliger, C. Bacterias versus fungal laccase: potential for micropollutant degradation. *AMB Express.* 2013, 3, 63, 1-14
78. Schlosser, D.; Grey, R.; Fritsche, W. Patterns of ligninolytic enzymes in *Trametes versicolor*. Distribution of extra and intracellular enzymes activities during cultivation on glucose, wheat straw and beech wood. *Appl. Microbiol. Biotechnol.* **1997**, 47, 412-418.
79. Cañas, A. I.; Camarero, S. Laccases and their natural mediators: Biotechnological tools for sustainable eco-friendly processes. *Biotechnol. Adv.* **2010**, 28, 694-705.
80. Valls, C.; Colom, J. F.; Baffert, C.; Gimbert, I.; Roncero, M. B.; Sigoillot, J.-C. Comparing the efficiency of the laccase-NHA and laccase-HBT system in eucalyptus pulp bleaching. *Biochem. Eng. J.* **2010**, 49, 401-407.
81. Weng, S. S.; Ku, K.-L.; Lai, H.-T. The implication of mediators for enhancement of laccase oxidation of sulfonamide antibiotics. *Bioresour. Technol.* **2012**, 113, 259-164.
82. Chen, B.-Y.; Chen, S.-Y.; Lin, M.-Y.; Chang, J.-S. Exploring bioaugmentation strategies for azo-dye decolorization using mixed consortium of *Pseudomonas luteola* and *Escherichia coli*. *Process Biochem.* **2006**, 1574-1581.
83. Kalyani, D. C.; Patil, P.S.; Jadhav, J. P.; Govindwar S.P. Biodegradation of reactive textile dye Red BLI by an isolated bacterium *Pseudomonas sp*. SUK1. *Bioresour Technol.* **2007**, 99: 4635-4641.
84. Kadam A. A.; Telke A. A.; Jagtap S. S.; Govindwar S. P. Decolorization of adsorbed textile dyes by developed consortium of *Pseudomonas sp.* SUK1 and *Aspergillus ochraceus* NCIM-1146 under solid state fermentation. J. Hazard. Mat. **2011**, 189: 486-494.
85. Phugare, S. S.; Kalyani, D. C.; Surwase, S. N.; Jadhav, J. P. Ecofriendly degradation, decolorization and detoxification of textile effluent by a developed bacterial consortium . *Ecotoxicol. Environ. Saf.* **2011**, 74, 1288-1296.
86. Barsing, P.; Tiwari, A.; Joshi, T.; Garg, S. Application of a novel bacterial consortium for mineralization of sulphonated aromatic amines. *Bioresour. Technol.* **2011**, 102, 765-771.
87. Oren, A.; Gurevich, P.; Henis, Y. Reduction of nitrosubstituted aromatic compounds by the halophilic anaerobic eubacteria *Haloanaerobium praevalens* and *Sporohalobacter marismortui*. *Appl. Environ. Microbiol.* **1991**, 57(11), 3367-3370.
88. Zeyer, J.; Kocher, H. P.; Timmis, K. N. Influence of para-Substituents on the Oxidative Metabolism of O-Nitrophenols by *Pseudomonas putida* B2. *Appl. Environ. Microbiol.* **1986**, 52 (2), 334-339.