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OPTIMIZATION OF MEDIA COMPONENTS AND PROCESS PARAMETERS FOR MICROBIAL MEDIATED REMEDIATION OF AZO DYES: A REVIEW

*Rajeswari Uppala and Azhaguchamy Muthukumaran**

Address(es): Dr. Azhaguchamy Muthukumaran,
Department of Biotechnology, Kalasalingam Academy of Research and Education, Krishnankoil 626126, Tamil Nadu, India, Phone Number: +91- 9486276120.

*Corresponding author: a.muthukumaran@klu.ac.in

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ABSTRACT

Azo dyes are one of the most commonly used synthetic dyes with enormous applications in the textile industry. The recalcitrant properties of azo dyes could be attributed to the highly complex chemical organization. The limitations such as high cost and emergence of secondary toxic pollutants as by-products associated with physicochemical mode of degradation urged researchers to explore potential alternatives. Microorganisms having versatile metabolic pathways and adaptations to different environmental conditions gained the attention of researchers to exploit them for azo dyes degradation in a cost-effective manner. The azo dye degradation using microbial sources proved to be a promising approach as compared to conventional physicochemical approaches. Microorganisms can induce different metabolic pathways in response to the external environment. The biodegradation efficacy of the microorganisms-based approach can be maximized by optimizing the culture media and process parameters. Optimization techniques predict the conditions required to increase the efficacy of azo dyes degradation by microbial sources and also decrease the number of experimental runs to achieve the maximum percentage of degradation. Interaction of variables such as medium components and process parameters can be determined using optimization tools. Response Surface Methodology (RSM) and Artificial Neural Network (ANN) based optimization approaches were discussed in this review with special emphasis on microbial degradation of azo dyes.

Keywords: Optimization, Media Components, Process Parameters, Remediation, Azo Dyes

INTRODUCTION

The world has reached a stage where the increase in population enhances the demand for the enhanced manufacturing of value-added products, including foods, inks, cosmetics, paper and textiles. Synthetic dyes serve as the primary raw materials in manufacturing these valuable products of day-to-day use. According to an estimation, the number of commercially available synthetic dyes exceeds over 0.1 million with whopping production of different dyestuff (approximately 7×10^5 tons) annually, which corresponds to the massive and indiscriminate industrialization policies (**Mohan *et al.*, 2004**). Among the synthetic dyes used in textile industries, the azo class of dyes, which are characterized by the presence of azo bonds (-N=N-), constitute the most predominant synthetic dyes in the form of diversity. Azo dyes contribute a significant share in providing environmental contaminants in the form of toxic effluents and bypass the remediation treatments owing to their molecular complexity (**Almeida & Corso, 2014**). The proportionate enhancement in the release of effluents in the form of wastewaters from textile industries owing to the indiscriminate application of synthetic dyes is the main cause of environmental pollution across the globe. Apart from the environmental effects, the extended stability of these synthetic dyes and by-products contributes to toxicity and mutagenicity in various life forms (**A. B. dos Santos *et al.*, 2007**).

Adverse effects of synthetic azo dyes

The technological expansion and massive industrialization process provide valuable products of day to day use at ease; however, the effluent released from these industries, especially from the textile industries, contains different types of heavy metals as well as a variety of synthetic dyes. Among these dyes, due to high molecular weight and complex structures, azo dyes are considered as most important owing to its low biodegradability, high toxicity, capacity to produce ecological/ environmental disturbances and severe health consequences (**Verma *et al.*, 2012**). Though the application of the azo dyes is widespread in food, pharmaceutical, textile, cosmetics and leather industries, its improper discharge to the environment followed by severe environmental consequences remains a critical issue to be taken care of (**Chakraborty *et al.*, 2013**). Even in low concentration, azo dyes present in the industrial effluent has a drastic influence on the aquatic ecosystem and severe health consequences in human being owing to its carcinogenic property (**Tan *et al.*, 2016**). In addition to that, the presence of the electron-deficient xenobiotic azo dyes in the industrial effluents has a characteristic influence on the development of mutagenesis, which ultimately affects the growth and development of human beings and other organisms (**Saratale *et al.*, 2011**; **R. L. Singh *et al.*, 2015**). For instance, the extensive use of azo dye, sunset yellow in food and packaging industries in the developed countries like the USA and Japan, leads to a detrimental effect on health owing to its severe cytotoxicity (**Dwivedi & Kumar, 2015**). The products of dye-based industries mainly include the generation of effluents in the form of wastewater, which possesses serious problems such as groundwater depletion and environmental deterioration associated with ecosystem services. Besides, the exhaustive use of the highly reactive azo dyes in the textile industries resulting in development of severe health consequences such as allergic dermatitis and bladder cancer (**Aravind *et al.*, 2016**). Owing to the toxicity shown by azo dyes in the textile-based industries, they are considered as emerging and significant environmental contaminants with a significant impact on the health of aquatic organisms as well as humans (**Ribeiro & Umbuzeiro, 2014**). The indiscriminate use of azo dyes in textile industries leads to the generation of effluents in the form of several intermediate and highly stable benzidine components, which have the inherent capacity of carcinogenicity with special reference to bladder cancer, prevalent in humans as well as animals (**Chung, 2016**). The xenobiotic and recalcitrant properties of azo dyes have a profound impact on human health, such as carcinogenic effects on the spleen and liver, chromosomal aberrations and nuclear anomalies (**Puvaneswari *et al.*, 2006**). In addition to the textile industries, the food and pharmaceutical industries are the second most prevalent users of these synthetic azo dyes for packaged food and pharmaceutical products. The indiscriminate use of the highly stable artificial azo dyes in different food-based industries leads to a severe loss of learning and memory function due to excessive brain tissue damage (**Gao *et al.*, 2011**). The rigorous application of azo dyes as food additives and cosmetics leads to severe health hazards, including asthma and other associated health problems (**Gil, 2014**).

Chemical remediation of azo dyes and its limitations

From the last few decades owing to the increase in urbanization and massive industrialization; an increase in the level of environmental pollutants and contaminants in the form of wastewater discharge from these industries has been observed. The indiscriminate use of the highly stable, recalcitrant azo dyes in these industries adversely affects not only the natural resources like soil fertility, aquatic biota but also affect the health of humans and ecosystem functioning. In this context, methods for proper reduction of these azo dyes from the wastewater effluents remain an uphill challenge (**Sudha & Saranya, 2014**). Owing to the complex structural varieties found in the available azo dyes, they are readily resistant to aerobic biodegradation as well as convention wastewater treatment technologies (**Huang *et al.*, 2015**). Conventional wastewater treatment technologies such as adsorption, coagulation/ flocculation, membrane filtration, electrolysis and ozonization are employed for the removal of azo dyes associated with industrial effluents. These conventional techniques proved to be inadequate for complete removal of azo dyes owing to the constraints such as inefficacy, limited applicability, requires further secondary treatment, high cost, generation of highly toxic waste materials as well as potential secondary pollutants and lack of environmental friendliness (**Asad *et al.*, 2007**; **Pandey *et al.*, 2007**; **Tahir *et al.*, 2016**). Besides, these conventional Physico-chemical based technologies are lagging behind to meet today's environmental conditions and demand for efficient dye removal and degradation from the industrial effluents (**Du *et al.*, 2015**). In addition to these constraints, these techniques also possess significant operational difficulties with the exploitation of more energy-intensive processes (**Krishnan *et al.*, 2017**). Among the different physical/chemical/biological techniques used in the remediation of these azo dyes from the wastewater effluents, biological processes have received considerable attention owing to several advantages such as cost-effective operational procedures with comparatively less amount of sludge during the process and eco-friendly nature (**Huang *et al.*, 2015**; **Lalnunhlimi & Krishnaswamy, 2016**). Among the approaches involved for bioremediation of azo dyes from industrial sludge, exploitation of microorganisms in decolourization and degradation process vows to be a cost effective, high efficiency and ecofriendly alternative to the available conventional strategies (**Deng *et al.*, 2008**; **Li *et al.*, 2015**).

Bacteria as a promising alternative to the remediation process

Owing to the constraints shown by the conventional physical and physicochemical wastewater treatment technologies in the removal of azo dyes efficiently; the focus is shifting towards the exploitation of biological sources, especially from microbial moieties due to their inherent and exceptional dye removal capacity. Microorganisms (bacteria, fungi, and yeast) have the inherent ability to synthesize a diverse group of enzymes with the potential to remove highly toxic azo dyes from the industrial effluents (**Corso & Maganha de Almeida, 2009**). Among the microbial sources, the bioremediation strategies mainly center around the exploitation of bacterial biomass and products owing to the cost-effectivity, ecofriendly nature and comparatively less production of sludge during the remediation process as compared to physical/ chemical or other microbial sources. Bacterial bioremediation strategies mainly occur through the process of biosorption or degradation through the action of bacterial enzymes or could involve both the abovementioned approaches for effective neutralization of hazardous synthetic azo dyes (**Solís *et al.*, 2012**). The efficacy of bacteria in azo dye decolorization/ degradation as compared to fungi can be attributed to its simplicity, cost-effectivity, a faster rate of decolorization and moreover inherent capacity to degrade the azo dyes reductively in an anaerobic condition (**Ali, 2010**; **Khalid *et al.*, 2008**). In this context, the exploitation of bacteria in the remediation of azo dyes from industrial effluents proves to be the most economical alternative strategy evolved to date.

Bioremediation of azo dyes by Gram + ve bacteria

The bioremediation or decolourization of highly toxic azo dyes from textile effluents by exploitation of microorganisms, especially bacteria is extensively studied in the present scenario of obtaining better remediation efficacy with any harsh environmental effect. In this context, *Bacillus* sp. is the most studied organism in remediation of textile azo dyes effectively as compared to other bacterial species (**Srinivasan *et al.*, 2014**). An important constituent of textile wastewater is crystal violet, which is an important member of azo dyes family and hence its remediation is highly necessary in terms of minimizing environmental deterioration and health hazards. Shah *et al.* (2013) successfully investigated the efficacy of *B. subtilis* ETL-2211 in remediation of toxic crystal violet from the textile effluents which basically depends upon the optimized nutritional and environmental parameters (**Shah *et al.*, 2013**). The efficacy of *Bacillus* sp. in remediation/ decolourization is not only limited to antharaquinone based dyes (Acid Blue) but also highly effective towards the decolourization of Malachite Green and Basic Blue X-GRRL (**Deng *et al.*, 2008**). From several decades, Indigo carmine is extensively exploited as an important dye in textile industries and strongly affect the health and also create environmental hazards. In this context, a cost effective and highly efficient remediation strategy should be applied to remediate highly toxic Indigo carmine from textile effluents. Li *et al.* (2015) successfully established the reductive decolourization of indigo carmine with the exploitation of *Bacillus* sp. MZS10 by virtue of its quinone dehydrogenase activity (**Li *et al.*, 2015**). In 2013, *Lysinibacillus* sp. KMK-A was successfully investigated to mitigate the azo bond removal from azo dye, Reactive Orange M2R from metal contaminated dye effluents in an eco-friendly manner (**Chaudhari *et al.*, 2013**). Apart from *Bacillus* sp., *Enterococcus faecalis* YZ 66 is successfully exploited for its effective bioremediation of highly toxic azo dye, Reactive Orange 16 (**Sahasrabudhe *et al.*, 2014**).

Bioremediation of azo dyes by Gram –ve bacteria

Degradation of synthetic azo dyes using chemicals will affect the ecological balance and also causes environmental problems and health hazards to human beings. Natural sources like microbes have a significant role in the removal of azo dyes from the environment. Degradation of Azo dyes using microorganisms is a most promising alternative to the conventional methods of bioremediation. Microorganisms show the key role in the degradation of synthetic azo dyes in an eco-friendly way. In this regard, the exploitation of Gram-negative bacteria and the enzymes produced by them in the remediation process is of prime importance. In this context, the enzyme i.e. laccase produced by *Pseudomonas putida* showed very promising bioremediation potential in remediating highly toxic synthetic azo dyes and other industrial effluents (**Kuddus *et al.*, 2013**). Besides, *Pseudomonas* sp., *Klebsiella* sp., and *Salmonella* sp. also contributed substantially to remediate highly toxic synthetic textile azo dye, Orange 3R, from the industrial effluent under optimized conditions (**Ponraj *et al.*, 2011**).

Factors affecting the bioremediation of azo dyes

Since the bacterial bioremediation process of many reactive dyes from the wastewater is comparatively faster and reliable than the fungal bioremediation process, considerable attention has been targeted towards the utilization of bacteria and bacterial-derived secondary metabolites for the bioremediation process. In addition, as complete mineralization and decolorization of reactive synthetic dyes depend upon the optimized conditions of cultural and nutritional parameters; formulating optimization parameters is highly important for the efficient remediation of the synthetic dyes as well as consistency in remediation (**Kumar Garg *et al.*, 2012**; **Lone *et al.*, 2015**). Carbon and nitrogen sources are used as nutritional parameters, whereas temperature, pH, initial concentration of reactive dyes, incubation time, and agitation are used as process parameters for optimization (**Khan *et al.*, 2014**).

PROCESS OPTIMIZATION PARAMETERS AFFECTING BACTERIAL BIOREMEDIATION

The optimization of the bacterial remediation process depends upon two types of optimization parameters, such as process/environmental parameters and the other one is the nutritional/medium optimization parameters. The initial concentration of reactive dyes, temperature, pH, incubation time, agitation is regarded as the process optimization parameters.

Temperature

The incubation temperature critically determines the efficacy of bacterial bioremediation process as at different temperature range the growth and metabolites produced by concerned bacteria essentially differ which eventually affects the dye decolorization/ degradation process. Hassan *et al*. (2015) reported the optimized temperature for effective bioremediation of reactive red synozol, disperse yellow and disperse blue by *Klebsiella* spp. was 35 °C (**Hassan *et al.*, 2015**). Illakiam *et al.* (2016) also successfully investigated the optimized temperature range of 35-37°C for bioremediation of different synthetic dyes by the majority of bacteria. The results suggested the highest efficacy of *Escherichia coli* and *Pseudomonas* sp. in remediating Alizarin red S dye at the optimized temperature of 37 °C (**Illakkiam *et al.*, 2016**).

pH

The pH plays a pivotal role in maintaining the efficiency of dye decolorization, degradation and overall remediation process. Generally, the optimal pH for the majority of dye removal by bacteria is often ranged between 6.0 and 10.0 (**Lavanya *et al.*, 2014**). The textile industries utilize reactive azo dyes under alkaline conditions for high throughput productivity as these processes are directly dependent upon a different range of pH. It was evident that at an optimal pH range from 6-12, *Clostridium bifermentans* has the ability to completely decolorize Reactive Red 3B-A dye (**Bardi & Marzona, 2010**). Singh *et al.* (2014) also reported earlier about the optimized pH of 7.0 for *Staphylococcus hominis* RMLRT03 for decolorization of Acid Orange (**R. Singh *et al.*, 2014**).

The initial concentration of dyes

The rate and amount of decolorization, degradation/mineralization of reactive synthetic dyes by bacteria is highly dependent upon the initial dye concentration as it directly affects the dye removal process owing to the availability on the adsorbent surface (**Yagub *et al.*, 2014**). Ogugbue and Sawidis (2011) earlier reported the optimized initial concentration of Acid Red 249 to be 100 mg/L for effective remediation by *Bacillus firmus* (**Ogugbue & Sawidis, 2011**). This result was further supported by the report given by Krishnan *et al*. (2017), where the results suggested that there was a marked decrease in the degradation efficacy when Brilliant Red X-3B, Direct Blue-6 and Direct Black-19 were used above 100 mg/L (**Krishnan *et al.*, 2017**).

Agitation

The dye remediation capacity of bacteria mainly depends upon the agitation parameters as it directly/indirectly correlates with the oxygen requirement of bacteria. N. Arunagirinathan *et al.* (2017) recently reported the bioremediation efficacy of *E. coli* AKIP-2 in remediating Evan Blue dye under the optimized condition of agitation. The results suggested that the bacterial remediation of Evan Blue was maximized under static condition and the efficacy decreases with the increase in the agitation speed (**N. Arunagirinathan *et al.*, 2017**). Similar results were obtained before, where three bacterial isolates, namely *P. aeruginosa*, *P. putida* and *B. cereus* attained maximized (90-94%) dye remediation of an array of synthetic dyes such as Acid Red-151, Orange II, Sulfur Black and Drimarene Blue under static condition (**Bayoumi *et al.*, 2014**).

Incubation time

The incubation period also plays a critical role in the bacterial bioremediation process. The highest efficacy of *Bacillus* sp. in remediating Acid Red 2 and Acid Orange 7 was observed with an optimized incubation period of 72 and 48 h, respectively. The results suggested that under an optimized incubation period, the efficacy of remediation varies from organism to organism as well as for different dyes (**Jaiswal & Gomashe, 2017**). The wide range of incubation periods was optimized for different bacteria targeting different reactive dyes, as reported earlier (**Rajan *et al.*, 2013**).

Medium optimization parameters affecting bacterial bioremediation

Carbon sources

Along with the process parameters, nutritional/medium parameters optimization also plays a critical role in reactive dyes remediation by bacteria. Among the nutritional parameters, carbon sources are essential for the bacterial bioremediation process. Ebency et al. (2013) reported that *Bacillus* sp. efficiently remediate reactive dye, Indigo Blue, with an efficacy of 86.25% with sucrose as the sole carbon source (**Ebency *et al.*, 2013**). Bheemaraddi *et al.* (2014) also reported that *P. aeruginosa* GSM3 effectively remediate the azo textile dye, Reactive violet 5 under different carbon sources, with glucose being the most efficient carbon source with 100% remediation within 24h of incubation as compared to sucrose which attains maximum efficacy at 26h (**Bheemaraddi *et al.*, 2014**).

Nitrogen sources

Nitrogen sources also interfere with the efficacy of decolorization and degradation of toxic azo dyes by bacteria. Gomaa, 2016 reported that four bacterial isolates such as *B. subtilis*, *B. cereus*, *B. licheniformis* and *Pseudomonas* sp. effectively remediate Black B and Congo red when peptone and yeast extract were used as optimized nitrogen sources (**Gomaa, 2016**). Earlier investigations also suggested the efficacy of bacterial bioremediation of highly reactive RB5 dye using yeast extract as the sole nitrogen source (**Johari, 2014**).

MICROBIAL ENZYME MEDIATED AZO DYES DEGRADATION

Microbial degradation of dyes involves different intracellular and extracellular enzyme systems. The enzymatic mode of azo dyes degradation is brought about by azoreductase, laccases, hydroxylases and peroxidase. Laccases and azoreductase have the potential to decolorize synthetic dyes of different chemical class (**R. L. Singh *et al.*, 2015**). Fungal enzymes also have the potential to oxidize a series of dyes due to their non-specificity towards dyes with varying structural conformations. The fungal enzymes such as peroxidase, laccase, manganese peroxidase and tyrosinase characteristically degrade textile dyes. On the other hand, bacterial biodegradation of dyes is generally associated with azo reductase, DCIP-reductase and laccase (**H. S. Lade *et al.*, 2012**).

For example, *B. laterosporus* exhibited 100% decolorization of DR54 within 48 h of incubation under optimized conditions with an increase in the enzymatic activities of tyrosinase, veratrine alcohol oxidase and NADH–-DCIP reductase (**Kurade *et al.*, 2016**). *Bacillus circulans* BWL1061 decolorized methyl orange due to increased activity of azoreductase, NADH-DCIP reductase, and laccase (**Liu *et al.*, 2017**). Salt-tolerant yeast *Pichia occidentalis* exhibited 98% decolorization of Acid Red B (ARB) with the involvement of NADH-DCIP reductase followed lignin peroxidase, manganese peroxidase and laccase (**Song *et al.*, 2017**).

Recently, consortial approaches have been gaining much interest in the remediation of textile dyes. In this system, the combined effects of various enzymes significantly enhances the dye degradation efficacy as compared to individual cultures. A consortium of *Aspergillus ochraceus* NCIM-1146 and *Pseudomonas* sp. SUK1 was reported for their ability to enhance dye decolorization of Rubine GFL to 95% in 30 h as compared to 46 and 63% decolorization when *A. ochraceus* NCIM-1146 and *Pseudomonas* sp. SUK1 was taken separately. The promising results could be attributed to the enhanced activity of laccase, veratryl alcohol oxidase, azo reductase and NADH-DCIP reductase. In another report, the bioremediation of Rubine GFL using a consortium of *Galactomyces geotrichum* MTCC 1360 and *Brevibacillus laterosporus* MTCC 2298 achieved 100% decolorization due to the activation of laccase, veratryl alcohol oxidase, tyrosinase, azo reductase, and riboflavin reductase (**Waghmode *et al.*, 2012**).

As presented in Table 1, microorganisms based reductive and oxidative enzymes are highly influential in the process of bioremediation. The complete degradation of azo dyes includes anaerobic decolorization in presence of flavin-dependent and flavin-independent azoreductases followed by an oxidative process in presence of peroxidases, laccases and tyrosinases (**Mahmood *et al.*, 2015**). In a report of Lade *et al.* (2015) a bacterial consortium constituting of *Providencia rettgeri* HSL1 and *Pseudomonas* sp. SUK1 exhibited 98-99 % decolorization of Reactive Black 5, Reactive Orange 16, Disperse Red 78 and Direct Red 81. The promising results could be attributed to the enhanced activity of azoreductase and NADH-DCIP reductase in the cleavage of complex azo interactions. Further, laccase and veratryl alcohol oxidase were reported for oxidation of toxic amines which are formed in the process (**H. Lade *et al.*, 2015**).

Azoreductases

The bacterial membrane is inhabited by azoreductases known for cleaving the azo bonding using NADH or NADPH or $FADH_2$ as an electron donor (**Kurade *et al.*, 2016**). Under the action of azoreductases, the azo bridge cleaves, resulting in two arylamines that are usually toxic and carcinogenic in nature. Fortunately, laccase acts upon such amines and transforms them into their corresponding quinones and non-toxic by-products (**Zucca *et al.*, 2016**). These enzymes are oxygen sensitive and thus significantly inhibited by oxygen during the reduction mechanism (**Sudha & Saranya, 2014**). Karatay *et al.* (2015) investigated removing azo dye, Remazol Blue using *Bacillus megaterium*, *Micrococcus luteus* and *Bacillus pumilus*. The study revealed an increase in azoreductase activity by 39.9 U/mL for *B. pumilus* (**Karatay *et al.*, 2015**).

Peroxidases

Dye-decolorizing peroxidases are microbial hemoproteins that possess high substrate specificity and are known to successfully degrade azo dyes in the presence of hydrogen peroxide (**A. Santos *et al.*, 2014**). Peroxidases are predominantly synthesized by fungal species during the process of dyes degradation. Fungal peroxidase isolated from *Bjerkandera adusta* efficiently decolorized azo dye present in industrial effluent (**Baratto *et al.*, 2015**). Santos *et al.* (2014) identified two new bacterial dye-decolorizing peroxidases from *B. subtilis* and *P. putida* MET94. According to a report of Min *et al.* (2015), the peroxidase produced by *B. subtilis* KCTC2023 efficiently decolorize Reactive Blue19 and Reactive Black 5 (**Min *et al.*, 2015**).

Tyrosinases

Tyrosinases are tetramer enzymes containing four copper atoms per molecule and binding sites for two aromatic compounds and oxygen. Similar to laccases, this class of phenol oxidases catalyzes the oxidation of aromatic compounds without the presence of cofactors. This enzyme could work on a number of substrates (**Sudha & Saranya, 2014**). Franciscon *et al.* (2003) reported the influence of tyrosinase in the remediation of Reactive Yellow 107, Reactive Black 5, Reactive Red 198 and Direct Blue 71 by *Brevibacterium* sp. VN-15 (**Franciscon *et al.*, 2012**).

Laccases

Laccase is a low molecular weight, copper-containing polyphenol oxidases found in plants, insects, bacteria and fungi (**Yan *et al.*, 2014**). Laccases have the inherent potential to oxidize a wide variety of aromatic compounds due to non-specific oxidation capacity, non-requirement of cofactors and ability to use readily available oxygen using Cu^{2+} as the mediator. They have been studied extensively for their oxidizing effect towards various dyes (**Phugare *et al.*, 2011**). Laccases oxidize the azo dye to generate a phenoxy radical which is subsequently re-oxidized to produce carbonium ion by cross-coupling of the

reactive species, including the formation of C-C and C-O bonds between phenolic molecules and formation of C-N and N-N bonds between aromatic amines (**R. L. Singh *et al.*, 2015**). White-rot fungi particularly *Trametes* sp. are the predominant source for laccases with characteristic features such as resistance to high alkalinity, extreme acidity, organic solvents, heavy metals and high thermal stability. Hence, *Trametes* sp. derived laccases have gained considerable attention (**Yan *et al.*, 2014**). Several laccase-producing fungal cultures were reported for the degradation of azo dyes (Table 1). However, high temperature and alkaline conditions are the limitations associated with fungal-derived laccase (**R. L. Singh *et al.*, 2015**; **Sudha & Saranya, 2014**).

Depending on the species and environmental conditions, fungal laccases are often secreted extracellularly in the form of different isoenzymes (**R. L. Singh *et al.*, 2015**). According to He *et al.* (2014), three laccase isoenzymes were purified from *Ganoderma* sp. En3. The isoenzymes exhibited promising decolorization ability. However, the enzymatic decolorization of dyes was efficiently enhanced when different laccase isoenzymes were used in combination due to their synergistic effect (**He *et al.*, 2015**).

The laccase activity is also influenced by the media composition and the conditions of fermentation. Metal ions such as copper and manganese also regulate the expression of regulatory genes encoding laccase isoenzymes (**He *et al.*, 2015**). Jiang *et al.* (2013) reported the activation of laccase isoenzyme produced by *Coprinus comatus* and its dye decolorization efficacy. The production of laccase isoenzyme and the enzymatic activity were influenced by the C/N ratio, aromatic compounds and copper content. At the optimal conditions of high-nitrogen and low-carbon, *C. comatus* produced six laccase isoenzymes with an efficiency of more than 90% when crude laccase was used to remediate Reactive Brilliant Blue K-3R, Reactive Dark Blue KR, and Malachite Green (**Jiang *et al.*, 2013**). Zhuo *et al.* (2017) reported the synergistic effect of Fe^{2+} and Cu^{2+} ions and aromatic compounds (vanillic acid, cinnamic acid, and ferulic acid) on the increased production of extracellular laccase in *P. ostreatus* HAUCC 162. In addition, the crude laccase significantly decolorized Methyl orange (**Zhuo *et al.*, 2017**).

Table 1 Microbial enzyme mediated degradation of azo dyes.

Enzymes	Organism	Dye	Reference
Manganese peroxidase	Phanerochaete sordid YK-624	Reactive Red 120	(**Harazono *et al.*, 2003**)
Peroxidase, Laccase, and Azoreductase	*Exiguobacterium* sp. RD3	Reactive blue 172	(**Dhanve *et al.*, 2008**)
Laccase	*Pseudomonas* sp. SU-EBT	Congo red	(**Telke *et al.*, 2010**)
Laccase, Veratryl alcohol oxidase, Azo reductase and NADH-DCIP reductase	*A. ochraceus* NCIM-1146 and *Pseudomonas* sp. SUK1	Rubine GFL	(**H. S. Lade *et al.*, 2012**)
Tyrosinase, Veratryl alcohol oxidase and NADH–-DCIP reductase	*Brevibacillus laterosporus*	Disperse Red 54	(**Kurade *et al.*, 2016**)
NADH-DCIP reductase, Peroxidase, Manganese peroxidase and Laccase	*Pichia occidentalis*	Acid Red B	(**Song *et al.*, 2017**)
Laccase and Reductase	*Pseudomonas* species	Reactive Orange 16	(**J. P. Jadhav *et al.*, 2010**)
Azo reductase, NADH-DCIP reductase, Veratryl alcohol oxidase and Tyrosinase	*Brevibacillus laterosporus*	Remazol red and Rubine GFL	(**Kurade *et al.*, 2013**)
Azoreductase, Lignin peroxidase and Laccase	*Sphingomonas paucimobilis, B. cereus* ATCC14579, *B. cereus* ATCC11778	Methyl orange	(**Ayed *et al.*, 2010**)
Alcohol oxidase	*P. aeruginosa* BCH	Remazol Black	(**Phugare *et al.*, 2011**)
Alcohol oxidase	*Comamonas* UVS	Red HE7B and Direct Blue GLL	(**U. U. Jadhav *et al.*, 2009**)
Azoreductase and NADH-DCIP reductase	*Providencia rettgeri* HSL1 and *Pseudomonas sp*. SUK1	Reactive Black 5 (RB 5), Reactive Orange 16 (RO 16), Disperse Red 78 (DR 78) and Direct Red 81 (DR 81)	(**H. Lade *et al.*, 2015**)
Laccase, Veratryl alcohol oxidase, Tyrosinase, Azo reductase, And Riboflavin reductase	*G. geotrichum* MTCC 1360 and *B. laterosporus* MTCC 2298	Rubine GFL	(**Waghmode *et al.*, 2012**)
Laccase	*Trametes trogii* S0301	Malachite green, Bromophenol blue, Crystal violet and acid Red	(**Yan *et al.*, 2014**)

Response surface methodology (RSM) for Optimization of azo dyes remediation

The dye remediation process is governed under the influence of numerous factors and their combined effect of which directly determines the process efficiency and performance of the designed system. Hence, for prediction and optimization of the process variables, different experimental models were designed and developed statistically (**Witek-Krowiak *et al.*, 2014**). The optimization process aims to identify the specific set of parameters that will result in the best possible outcome. Usually, for determining the effect of process variables, the One Variable at Time (OVAT) method is used where the independent variable is systemically changed while keeping the other parameters constant. However, this method is costly and time-consuming as all process variables are screened independently. Further, OVAT cannot provide any details on the interactions between the selected variables (**Kaur *et al.*, 2015**).

Hence, in the quest for experimental models to optimize the different variables in a multivariable system, RSM and Artificial Neural Network (ANN) are gaining much popularity as powerful data modeling tool[82]. The statistical design of experiments (DOEs) associated with RSM experimental models has the inherent potential to define complex non-linear interactions between different independent variables and the resulting responses (**Kaur *et al.*, 2015**). In recent times, process parameters including initial dye concentration, pH, temperature and inoculum size are optimized using the RSM tool for efficient decolorization/degradation of synthetic dyes. One of the advantages of employing RSM is the determination of the effect of independent variables on the interactions of process parameters with a minimum number of experimental runs (**Senthilkumar *et al.*, 2012**). Hence, RSM improves process performance, reduces operation costs and experimental time (**Witek-Krowiak *et al.*, 2014**). Maqbool *et al.* (2016) reported the optimum salt content, pH, carbon content, concentration of metal mixtures using the RSM tool for determining the efficacy of *P. aeruginosa* ZM130 in decolorizing reactive red-120 (**Maqbool *et al.*, 2016**).

Several RSM designs have been developed and employed to optimize the biosorption process. As presented in Table 2, central composite design (CCD), Box–Behnken design (BB) and Plackett–Burman (PB) design have been widely employed to optimize numerous parameters associated with the decolorization of dyes. Design Expert (Stat-Ease, Inc.), Minitab (Minitab Inc.), Statistica (StatSoft), JMP (SAS) and Matlab (MathWorks) are widely used to study RSM based optimization of parameters in remediation of synthetic dyes. The response obtained in the form of 3D-graph and/or contour plot serve as a fast way of modelling when the optimal response is within experimental boundaries (**Witek-Krowiak *et al.*, 2014**).

Central composite design (CCD)

For high-quality predictions on linear and quadratic interaction effects of variables, CCD is widely used as a promising statistical design. The CCD constitutes fractional factorial design at two levels (2^n), center points (cp), which corresponds to the middle level of the factors, and axial points ($2n$) (**Witek-Krowiak *et al.*, 2014**). *Dietzia* sp. PD1 biodegraded Congo red and indigo and two levels three-factor (2^3) CCD was employed for optimization of pH, initial dye concentration and incubation time. At the optimized levels, the biodegradation efficacy for Congo red and indigo carmine was observed to be 99.97 and 99.95%, respectively (**P. Das *et al.*, 2016**). Hafshejani *et al.* (2013) reported the decolorization and degradation of Direct Blue 71 by *P. aeruginosa* with the three-level CCD to optimize different variables. At the optimal conditions of 35 °C, pH 8.0 and 49.9 mg/L initial dye concentration, the decolorization efficacy was observed to be 84.80 % (**Hafshejani *et al.*, 2014**). In a report by Senthilkumar *et al.* (2012), three-level CCD was employed to

optimize initial dye concentrations, carbon source and nitrogen source for efficient decolorization of Remazol Turquoise Blue (RTB) and Reactive Black 5 (RB5) using *Pseudomonas* sp. (**Senthilkumar *et al.*, 2012**). In a similar experiment, the effect pH, incubation time, and concentration of dye on decolorization efficacy of *Cordyceps militaris* were determined using CCD model (**Kaur *et al.*, 2015**). Yan *et al.* (2014) reported enhanced laccase production in *T. trogii* S0301 using CCD of RSM. On process optimization, the maximum laccase activity was attained at an optimum pH of 3.0 and temperature of 45 °C. Further, the purified laccase was found to significantly decolorized malachite green, bromophenol blue, crystal violet and acid red (**Yan *et al.*, 2014**).

Box–Behnken design (BB)

Box and Behnken design (BB) is a 3-level incomplete factorial design developed to minimize the number of experiments and extensively used in the optimization of numerous factors involved in dye removal. In BB design, The experiment matrices are constructed by means of two-level factorial designs (+1, −1) with incomplete block designs (**Witek-Krowiak *et al.*, 2014**). Garg *et al*. (2015) reported bioremediation of Reactive Orange 4 using *Pseudomonas putida* SKG-1. As indicated by the RSM-based BB design, the 97.8% decolorization was achieved at an optimized dye concentration of 50 mg/L, sucrose 0.7%, and peptone 0.28% upon 72 h of incubation (**Garg & Tripathi, 2017**). RSM-based BB design was employed by Das & Mishra (2017) in order to optimize the process parameters for efficient removal of Reactive Green-19 using bacterial consortium. The dye removal efficacy was observed to be 97% at the optimized temperature of 32 °C, pH 8.3 and Yeast Extract concentration of 1.16 g/100 mL (**A. Das & Mishra, 2017**). Sathian *et al.* (2013) also utilized BB design in order to optimize the levels of pH, temperature, agitation speed and dye concentration for determining the efficacy of *Pleurotus floridanus* in treatment of textile dye wastewater (**Sathian *et al.*, 2013**). The decolorization of Solophenyl red 3BL (SR) by *Fomes fomentarius* laccase was studied using RSM-based BB. The results indicated the optimal conditions with enzyme concentration of 0.8 U/mL, mediator concentration of 33 μM, and time of 14 h 30 min. The predicted optimal conditions resulted in 79.66% of decolorization, which significantly correlates with the predicted value of 80.70% (**Neifar *et al.*, 2011**).

Plackett–Burman design (PB)

The PB design was developed to determine the main factor effects for a process consisting of multiple variables in a short experimental time. In PB design, the number of experiments is equal to the number of parameters in the first order RSM model ($N = k + 1$), and the degree of freedom is equal to zero (**Witek-Krowiak *et al.*, 2014**). Hema and Suresha (2015) evaluated the ability of *Penicillium oxalicum* RF3 in decolorizing Isolan grey by employing RSM based PB design. At the predicted optimal parameters, an enhanced decolorization of Isolan grey was attained with maximum decolorization of 50.75% (**Hema & Suresha, 2015**).

Table 2 Optimization of bacterial remediation of azo dyes using Response surface methodology (RSM)

Organism	Dye	Parameters	RSM design	Reference
***Pseudomonas* sp.**	Congo red, Reactive red 195	initial dye concentration, carbon source, nitrogen source	central composite design (CCD)	(**Senthilkumar *et al.*, 2013**)
Pseudomonas aeruginosa	Direct Blue 71	temperature, medium pH, initial dye concentration	central composite design (CCD)	(**Hafshejani *et al.*, 2014**)
***Pseudomonas* sp.**	*Remazol Turquoise Blue, Reactive Black 5*	concentrations of Dye, Carbon source, Nitrogen source	central composite design (CCD)	(**Senthilkumar *et al.*, 2012**)
***Dietzia* sp. PD1**	Congo red, Indigo carmine	pH, initial dye concentration, time	central composite design (CCD)	(**P. Das *et al.*, 2016**)
Pseudomonas aeroginosa* PAO1 *Stenotrophomonas maltophila*, *Proteus mirabilis	Direct Black 22	Glucose concentration, yeast extract concentration, dye concentration, inoculum size	central composite design (CCD)	(**Mohana *et al.*, 2008**)
***Cordyceps militaris* MTCC3936**	Reactive yellow 18, Reactive red 31, Reactive black 8, Reactive green 19 Reactive red 74.	pH, incubation time, the concentration of dye	Centre composite rotatable design (CCRD)	(**Kaur *et al.*, 2015**)
***Trametes trogii* laccase**	Malachite green, Bromophenol blue, Crystal violet, Acid red	pH, temperature	central composite design (CCD)	(**Yan *et al.*, 2014**)
***Pseudomonas aeruginosa* BCH**	Remazol Orange	pH, temperature, cell mass concentration	Box–Behnken design	(**S. B. Jadhav *et al.*, 2013**)
***Pseudomonas putida* SKG-1**	Reactive orange 4	Dye concentration, sucrose, peptone, incubation time	Box–Behnken design	(**Garg & Tripathi, 2017**)
Bacillus subtilis	Disperse Yellow 211	temperature, pH and initial dye concentration	Box–Behnken design	(**Sharma *et al.*, 2009**)
Bacterial consortium	Reactive Green-19	pH, incubation temperature, Yeast extract concentration	Box–Behnken design	(**A. Das & Mishra, 2017**)
***Fomes fomentarius* laccase**	Solophenyl red 3BL	enzyme concentration, redox mediator concentration, incubation time	Box–Behnken design	(**Neifar *et al.*, 2011**)
***Penicillium oxalicum* RF3**	Isolan Grey	Inoculum size, media components, pH, temperature, dye concentration, incubation time	Plackett-Burman	(**Hema & Suresha, 2015**)

Optimization of bacterial remediation of azo dyes using artificial neural network (ANN)

Apart from RSM, ANN proved to be a valuable tool in modeling and optimization of variable parameters for efficient removal of dyes. The efficacy of ANN could be attributed to recognize and reproduce cause−effect relationships through evaluation for multiple input−output systems. The statistical aspect of ANN aids in determining the factors which have a significant effect on the biosorption process. Using ANN, the number of experiments needed in an experimental design and time can be significantly reduced (**Witek-Krowiak *et al.*, 2014**). ANN is useful in simulating and up-scaling complex biological processes even without the description of the phenomena involved in the process (**Ghaedi *et al.*, 2014**).

In a report of Khataee *et al*. (2010) the three-layered feed-forward back propagation ANN model was employed to predict the decolorization efficiency of *Chara* sp. towards Malachite Green (MG). The process parameters like temperature, pH, initial dye concentration, reaction time and amount of algae on the decolorization efficiency were studied. The findings indicated that ANN provides reasonable predictive performance (R2 = 0.970) (**Khataee *et al.*, 2010**). Yang *et al.* (2011) documented the ANN-based modeling for the biosorption of Acid Black 172 (AB) and Congo red (CR) using *Penicillium* YW 01. Initial dye concentration and temperature were observed to be the most influential parameters for biosorption process as per the ANN-based analysis (**Yang *et al.*, 2011**). Das *et al.* (2015) documented the correlation between the input process variables and output parameters for degradation of Congo red and indigo carmine using *Dietzia* sp. PD1 by utilizing the ANN model (**P. Das *et al.*, 2016**).

CONCLUSION

An overview of various methods being employed to design and optimize dye degradation/decolorization was described. The microbial degradation efficacy depends upon the optimized levels of nutrients, pH, temperature, oxygen. These nutritional parameters can be optimized to enhance bioremediation efficacy using numerous software-based algorithms. Microbial enzymes based decolorization potential was also thoroughly described. Enzymes such as azoreductase, laccases, peroxidase, and hydroxylases are highly important in enhancing the degradation

of azo dyes. Recently, the consortial approaches have been gaining much interest in the remediation of textile dyes as the combined effects of various enzymes enhance dye degradation compared to individual cultures. The exploitation of microbial biosorbents as an efficient remediation approach instead of conventional approaches was also described in detail. Further, the involvement of RSM and ANN-based statistical tools promising alternative to predict and optimize the different variables in order to increase the efficacy of bioremediation of dyes. Hence, microorganism-mediated remediation of azo dyes serves as an efficient, cost-effective and eco-friendly alternative to the conventional Physico-chemical process for efficient removal and degradation of azo dyes from the industrial effluents.

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REFERENCES

Ali, H. (2010). Biodegradation of Synthetic Dyes—A Review. Water, Air, & Soil Pollution, 213(1–4), 251–273. https://doi.org/10.1007/s11270-010-0382-4

Almeida, E. J. R., & Corso, C. R. (2014). 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, 112, 317–322. https://doi.org/10.1016/j.chemosphere.2014.04.060

Aravind, P., Selvaraj, H., Ferro, S., & Sundaram, M. (2016). An integrated (electro- and bio-oxidation) approach for remediation of industrial wastewater containing azo-dyes: Understanding the degradation mechanism and toxicity assessment. Journal of Hazardous Materials, 318, 203–215. https://doi.org/10.1016/j.jhazmat.2016.07.028

Asad, S., Amoozegar, M. A., Pourbabaee, A. A., Sarbolouki, M. N., & Dastgheib, S. M. M. (2007). Decolorization of textile azo dyes by newly isolated halophilic and halotolerant bacteria. Bioresource Technology, 98(11), 2082–2088. https://doi.org/10.1016/j.biortech.2006.08.020

Ayed, L., Khelifi, E., Jannet, H. Ben, Miladi, H., Cheref, A., Achour, S., & Bakhrouf, A. (2010). Response surface methodology for decolorization of azo dye Methyl Orange by bacterial consortium: Produced enzymes and metabolites characterization. Chemical Engineering Journal, 165(1), 200–208. https://doi.org/10.1016/j.cej.2010.09.018

Baratto, M. C., Juarez-Moreno, K., Pogni, R., Basosi, R., & Vazquez-Duhalt, R. (2015). EPR and LC-MS studies on the mechanism of industrial dye decolorization by versatile peroxidase from *Bjerkandera adusta*. Environmental Science and Pollution Research, 22(11), 8683–8692. https://doi.org/10.1007/s11356-014-4051-9

Bardi, L., & Marzona, M. (2010). Factors Affecting the Complete Mineralization of Azo Dyes. In Handbook of Environmental Chemistry (Vol. 5, Issue Part N, pp. 195–210). https://doi.org/10.1007/698_2009_50

Bayoumi, M. N., Al-wasify, R. S., & Hamed, S. R. (2014). Bioremediation of Textile Wastewater Dyes using Local Bacterial Isolates. International Journal of Current Miccrobiology and Applied Science, 3(12), 962–970.

Bheemaraddi, M., Shivannavar, C., & Gaddad, S. (2014). Effect of carbon and nitrogen sources on biodegradation of textile azo dye Reactive Violet 5 by *Pseudomonas aeruginosa* GSM3. Scholars Academic Journal of Biosciences (SAJB), 2(4), 285–289.

Chakraborty, S., Basak, B., Dutta, S., Bhunia, B., & Dey, A. (2013). Decolorization and biodegradation of Congo red dye by a novel white rot fungus *Alternaria alternata* CMERI F6. Bioresource Technology, 147, 662–666. https://doi.org/10.1016/j.biortech.2013.08.117

Chaudhari, A. U., Tapase, S. R., Markad, V. L., & Kodam, K. M. (2013). Simultaneous decolorization of reactive Orange M2R dye and reduction of chromate by *Lysinibacillus* sp. KMK-A. Journal of Hazardous Materials, 262, 580–588. https://doi.org/10.1016/j.jhazmat.2013.09.006

Chung, K.-T. (2016). Azo dyes and human health: A review. Journal of Environmental Science and Health, Part C, 34(4), 233–261. https://doi.org/10.1080/10590501.2016.1236602

Corso, C. R., & Maganha de Almeida, A. C. (2009). Bioremediation of Dyes in Textile Effluents by Aspergillus oryzae. Microbial Ecology, 57(2), 384–390. https://doi.org/10.1007/s00248-008-9459-7

Das, A., & Mishra, S. (2017). Removal of textile dye reactive green-19 using bacterial consortium: Process optimization using response surface methodology and kinetics study. Journal of Environmental Chemical Engineering, 5(1), 612–627. https://doi.org/10.1016/j.jece.2016.10.005

Das, P., Banerjee, P., Zaman, A., & Bhattacharya, P. (2016). Biodegradation of two Azo dyes using *Dietzia* sp. PD1: process optimization using Response Surface Methodology and Artificial Neural Network. Desalination and Water Treatment, 57(16), 7293–7301. https://doi.org/10.1080/19443994.2015.1013993

Deng, D., Guo, J., Zeng, G., & Sun, G. (2008). Decolorization of anthraquinone, triphenylmethane and azo dyes by a new isolated *Bacillus cereus* strain DC11. International Biodeterioration & Biodegradation, 62(3), 263–269. https://doi.org/10.1016/j.ibiod.2008.01.017

Dhanve, R. S., Shedbalkar, U. U., & Jadhav, J. P. (2008). Biodegradation of diazo reactive dye Navy blue HE2R (Reactive blue 172) by an isolated *Exiguobacterium* sp. RD3. Biotechnology and Bioprocess Engineering, 13(1), 53–60. https://doi.org/10.1007/s12257-007-0165-y

dos Santos, A. B., Cervantes, F. J., & van Lier, J. B. (2007). Review paper on current technologies for decolourisation of textile wastewaters: Perspectives for anaerobic biotechnology. Bioresource Technology, 98(12), 2369–2385. https://doi.org/10.1016/j.biortech.2006.11.013

Du, L.-N., Li, G., Zhao, Y.-H., Xu, H.-K., Wang, Y., Zhou, Y., & Wang, L. (2015). Efficient metabolism of the azo dye methyl orange by *Aeromonas* sp. strain DH-6: Characteristics and partial mechanism. International Biodeterioration & Biodegradation, 105, 66–72. https://doi.org/10.1016/j.ibiod.2015.08.019

Dwivedi, K., & Kumar, G. (2015). Genetic Damage Induced by a Food Coloring Dye (Sunset Yellow) on Meristematic Cells of *Brassica campestris* L. Journal of Environmental and Public Health, 2015, 1–5. https://doi.org/10.1155/2015/319727

Ebency, C. I. L., Rajan, S., Murugesan, A. G., Rajesh, R., & Elayarajah, B. (2013). Biodegradation of textile azo dyes and its bioremediation potential using seed germination efficiency. International Journal of Current Microbiology and Applied Sciences, 2(10), 496–505.

Franciscon, E., Grossman, M., Paschoal, J. A., Reyes, F. G., & Durrant, L. (2012). Decolorization and biodegradation of reactive sulfonated azo dyes by a newly isolated Brevibacterium sp. strain VN-15. SpringerPlus, 1(1), 37. https://doi.org/10.1186/2193-1801-1-37

Gao, Y., Li, C., Shen, J., Yin, H., An, X., & Jin, H. (2011). Effect of Food Azo Dye Tartrazine on Learning and Memory Functions in Mice and Rats, and the Possible Mechanisms Involved. Journal of Food Science, 76(6), T125–T129. https://doi.org/10.1111/j.1750-3841.2011.02267.x

Garg, S. K., & Tripathi, M. (2017). Microbial Strategies for Discoloration and Detoxification of Azo Dyes from Textile Effluents. Research Journal of Microbiology, 12(1), 1–19. https://doi.org/10.3923/jm.2017.1.19

Ghaedi, M., Ghaedi, A. M., Negintaji, E., Ansari, A., & Mohammadi, F. (2014). Artificial neural network – Imperialist competitive algorithm based optimization for removal of sunset yellow using Zn(OH)2 nanoparticles-activated carbon. Journal of Industrial and Engineering Chemistry, 20(6), 4332–4343. https://doi.org/10.1016/j.jiec.2014.01.041

Gil, C. (2014). Toxicological effects of food additives – Azo dyes. SLU, Dept. of Biomedical Sciences and Veterinary Public Health, 1–22.

Gomaa, E. Z. (2016). Biodegradation and Detoxification of Azo Dyes by Some Bacterial Strains. Microbiology Journal, 6(1), 15–24. https://doi.org/10.3923/mj.2016.15.24

Hafshejani, M. K., Ogugbue, C. J., & Morad, N. (2014). Application of response surface methodology for optimization of decolorization and mineralization of triazo dye Direct Blue 71 by *Pseudomonas aeruginosa*. 3 Biotech, 4(6), 605–619. https://doi.org/10.1007/s13205-013-0192-7

Harazono, K., Watanabe, Y., & Nakamura, K. (2003). Decolorization of Azo Dye by the White-Rot Basidiomycete *Phanerochaete sordida* and by Its Manganese Peroxidase. Journal of Bioscience and Bioengineering, 95(5), 455–459. https://doi.org/10.1263/jbb.95.455

Hassan, S. E., Fouda, A., Azab, M. S., & Saied, E. (2015). International Journal of Environmental Biology Original Article Biological decolorization of different azo dyes using two bacterial strains of *Klebsiella* spp . and their consortium. International Journal of Environmental Biology, 5(4), 104–114.

He, F., Qin, X., Zhang, H., Yang, Y., Zhang, X., & Yang, Y. (2015). Characterization of laccase isoenzymes from the white-rot fungus *Ganoderma* sp.En3 and synergistic action of isoenzymes for dye decolorization. Journal of Chemical Technology & Biotechnology, 90(12), 2265–2279. https://doi.org/10.1002/jctb.4543

Hema, N., & Suresha, S. (2015). Optimization of Fungal Decolorization of Isolan Grey using Plackett-Burman Experimental Design. Research in Chemistry and Environment, 5(2), 26–34.

Huang, G., Wang, W., & Liu, G. (2015). Simultaneous chromate reduction and azo dye decolourization by *Lactobacillus paracase* CL1107 isolated from deep sea sediment. Journal of Environmental Management, 157, 297–302. https://doi.org/10.1016/j.jenvman.2015.04.031

Illakkiam, D., Subha, D., Ahila, V., & Geetha, N. (2016). Decolorization of alizarin red s dye by bacterial strains isolated from industrial effluents. International Journal of Plant, Animal and Environmental Sciences, 1, 268–276.
Jadhav, J. P., Kalyani, D. C., Telke, A. A., Phugare, S. S., & Govindwar, S. P. (2010). Evaluation of the efficacy of a bacterial consortium for the removal of color, reduction of heavy metals, and toxicity from textile dye effluent. Bioresource Technology, 101(1), 165–173. https://doi.org/10.1016/j.biortech.2009.08.027
Jadhav, S. B., Surwase, S. N., Phugare, S. S., & Jadhav, J. P. (2013). Response surface methodology mediated optimization of Remazol Orange decolorization in plain distilled water by *Pseudomonas aeruginosa* BCH. International Journal of Environmental Science and Technology, 10(1), 181–190. https://doi.org/10.1007/s13762-012-0088-9
Jadhav, U. U., Dawkar, V. V., Tamboli, D. P., & Govindwar, S. P. (2009). Purification and characterization of veratryl alcohol oxidase from *Comamonas* sp. UVS and its role in decolorization of textile dyes. Biotechnology and Bioprocess Engineering, 14(3), 369–376. https://doi.org/10.1007/s12257-008-0300-4
Jaiswal, S. S., & Gomashe, A. V. (2017). Bioremediation of textile azo dyes by newly isolated *Bacillus* sp. from dye contaminated soil. International Journal of Biotechnology and Biochemistry, 13(2), 147–153.
Jiang, M., Ten, Z., & Ding, S. (2013). Decolorization of Synthetic Dyes by Crude and Purified Laccases from *Coprinus comatus* Grown Under Different Cultures: The Role of Major Isoenzyme in Dyes Decolorization. Applied Biochemistry and Biotechnology, 169(2), 660–672. https://doi.org/10.1007/s12010-012-0031-z
Johari, W. L. W. (2014). Biodecolorization of Azo Dyes by Microorganisms Isolated From Serdang and Merambong Soils. Bioremediation Science and Technology Research, 2(1 SE-Articles).
Karatay, S. E., Kılıç, N. K., & Dönmez, G. (2015). Removal of Remazol Blue by azoreductase from newly isolated bacteria. Ecological Engineering, 84, 301–304. https://doi.org/10.1016/j.ecoleng.2015.09.037
Kaur, B., Kumar, B., Garg, N., & Kaur, N. (2015). Statistical Optimization of Conditions for Decolorization of Synthetic Dyes by *Cordyceps militaris* MTCC 3936 Using RSM. BioMed Research International, 2015, 1–17. https://doi.org/10.1155/2015/536745
Khalid, A., Arshad, M., & Crowley, D. E. (2008). Accelerated decolorization of structurally different azo dyes by newly isolated bacterial strains. Applied Microbiology and Biotechnology, 78(2), 361–369. https://doi.org/10.1007/s00253-007-1302-4
Khan, Z., Jain, K., Soni, A., & Madamwar, D. (2014). Microaerophilic degradation of sulphonated azo dye – Reactive Red 195 by bacterial consortium AR1 through co-metabolism. International Biodeterioration & Biodegradation, 94, 167–175. https://doi.org/10.1016/j.ibiod.2014.07.002
Khataee, A. R., Dehghan, G., Ebadi, A., Zarei, M., & Pourhassan, M. (2010). Biological treatment of a dye solution by *Macroalgae Chara* sp.: Effect of operational parameters, intermediates identification and artificial neural network modeling. Bioresource Technology, 101(7), 2252–2258. https://doi.org/10.1016/j.biortech.2009.11.079
Krishnan, J., Arvind Kishore, A., Suresh, A., Madhumeetha, B., & Gnana Prakash, D. (2017). Effect of pH, inoculum dose and initial dye concentration on the removal of azo dye mixture under aerobic conditions. International Biodeterioration & Biodegradation, 119, 16–27. https://doi.org/10.1016/j.ibiod.2016.11.024
Kuddus, M., Joseph, B., & Wasudev Ramteke, P. (2013). Production of laccase from newly isolated Pseudomonas putida and its application in bioremediation of synthetic dyes and industrial effluents. Biocatalysis and Agricultural Biotechnology, 2(4), 333–338. https://doi.org/10.1016/j.bcab.2013.06.002
Kumar Garg, S., Tripathi, M., Singh, S. K., & Tiwari, J. K. (2012). Biodecolorization of textile dye effluent by *Pseudomonas putida* SKG-1 (MTCC 10510) under the conditions optimized for monoazo dye orange II color removal in simulated minimal salt medium. International Biodeterioration & Biodegradation, 74, 24–35. https://doi.org/10.1016/j.ibiod.2012.07.007
Kurade, M. B., Waghmode, T. R., Khandare, R. V., Jeon, B.-H., & Govindwar, S. P. (2016). Biodegradation and detoxification of textile dye Disperse Red 54 by *Brevibacillus laterosporus* and determination of its metabolic fate. Journal of Bioscience and Bioengineering, 121(4), 442–449. https://doi.org/10.1016/j.jbiosc.2015.08.014
Kurade, M. B., Waghmode, T. R., Tamboli, D. P., & Govindwar, S. P. (2013). Differential catalytic action of *Brevibacillus laterosporus* on two dissimilar azo dyes Remazol red and Rubine GFL. Journal of Basic Microbiology, 53(2), 136–146. https://doi.org/10.1002/jobm.201100402
Lade, H., Kadam, A., Paul, D., & Govindwar, S. (2015). Biodegradation and detoxification of textile azo dyes by bacterial consortium under sequential microaerophilic/aerobic processes. EXCLI Journal 201, 14, 158–174.
Lade, H. S., Waghmode, T. R., Kadam, A. A., & Govindwar, S. P. (2012). Enhanced biodegradation and detoxification of disperse azo dye Rubine GFL and textile industry effluent by defined fungal-bacterial consortium. International Biodeterioration & Biodegradation, 72, 94–107. https://doi.org/10.1016/j.ibiod.2012.06.001
Lalnunhlimi, S., & Krishnaswamy, V. (2016). Decolorization of azo dyes (Direct Blue 151 and Direct Red 31) by moderately alkaliphilic bacterial consortium. Brazilian Journal of Microbiology, 47(1), 39–46. https://doi.org/10.1016/j.bjm.2015.11.013
Lavanya, C., Dhankar, R., Chhikara, S., & Sheoran, S. (2014). Review Article Degradation of Toxic Dyes : A Review. International Journal of Current Microbiology and Applied Sciences, 3(6), 189–199.
Li, H.-X., Xu, B., Tang, L., Zhang, J.-H., & Mao, Z.-G. (2015). Reductive decolorization of indigo carmine dye with *Bacillus* sp. MZS10. International Biodeterioration & Biodegradation, 103, 30–37. https://doi.org/10.1016/j.ibiod.2015.04.007
Liu, W., Liu, C., Liu, L., You, Y., Jiang, J., Zhou, Z., & Dong, Z. (2017). Simultaneous decolorization of sulfonated azo dyes and reduction of hexavalent chromium under high salt condition by a newly isolated salt-tolerant strain *Bacillus circulans* BWL1061. Ecotoxicology and Environmental Safety, 141(March), 9–16. https://doi.org/10.1016/j.ecoenv.2017.03.005
Lone, T. A., Revathi, C., & Lone, R. A. (2015). Isolation of Dye Degrading Bacillus Species from the Soil near Dyeing Industry and Its Potential Application in Dye Effluent Treatment. American-Eurasian Journal of Toxicological Sciences, 7(3), 129–135. https://doi.org/10.5829/idosi.aejts.2015.7.3.94162
Mahmood, S., Khalid, A., Arshad, M., Mahmood, T., & Crowley, D. E. (2015). Detoxification of azo dyes by bacterial oxidoreductase enzymes. Critical Reviews in Biotechnology, 36(4), 1–13. https://doi.org/10.3109/07388551.2015.1004518
Maqbool, Z., Hussain, S., Ahmad, T., Nadeem, H., Imran, M., Khalid, A., Abid, M., & Martin-Laurent, F. (2016). Use of RSM modeling for optimizing decolorization of simulated textile wastewater by *Pseudomonas aeruginosa* strain ZM130 capable of simultaneous removal of reactive dyes and hexavalent chromium. Environmental Science and Pollution Research, 23(11), 11224–11239. https://doi.org/10.1007/s11356-016-6275-3
Min, K., Gong, G., Woo, H. M., Kim, Y., & Um, Y. (2015). A dye-decolorizing peroxidase from *Bacillus subtilis* exhibiting substrate-dependent optimum temperature for dyes and β-ether lignin dimer. Scientific Reports, 5(1), 8245. https://doi.org/10.1038/srep08245
Mohan, S. V., Bhaskar, Y. V., & Karthikeyan, J. (2004). Biological decolourisation of simulated azo dye in aqueous phase by algae Spirogyra species. International Journal of Environment and Pollution, 21(3), 211. https://doi.org/10.1504/IJEP.2004.004190
Mohana, S., Shrivastava, S., Divecha, J., & Madamwar, D. (2008). Response surface methodology for optimization of medium for decolorization of textile dye Direct Black 22 by a novel bacterial consortium. Bioresource Technology, 99(3), 562–569. https://doi.org/10.1016/j.biortech.2006.12.033
N. Arunagirinathan, S. A. K., J. Hemapriya, S. V., & Indra, V. (2017). Bioremediation and Detoxification of a Textile Azo Dye-Evans Blue by Bacterial Strain AKIP-2. International Journal of Current Microbiology and Applied Sciences, 6(4), 2687–2694. https://doi.org/10.20546/ijcmas.2017.604.313
Neifar, M., Jaouani, A., Kamoun, A., Ellouze-Ghorbel, R., & Ellouze-Chaabouni, S. (2011). Decolorization of Solophenyl Red 3BL Polyazo Dye by Laccase-Mediator System: Optimization through Response Surface Methodology. Enzyme Research, 2011(1), 1–8. https://doi.org/10.4061/2011/179050
Ogugbue, C. J., & Sawidis, T. (2011). Optimisation of process parameters for bioreduction of azo dyes using Bacillus firmus under batch anaerobic condition. International Journal of Environmental Studies, 68(5), 651–665. https://doi.org/10.1080/00207233.2011.578353
Pandey, A., Singh, P., & Iyengar, L. (2007). Bacterial decolorization and degradation of azo dyes. International Biodeterioration & Biodegradation, 59(2), 73–84. https://doi.org/10.1016/j.ibiod.2006.08.006
Phugare, S. S., Waghmare, S. R., & Jadhav, J. P. (2011). Purification and characterization of dye degrading of veratryl alcohol oxidase from *Pseudomonas aeruginosa* strain BCH. World Journal of Microbiology and Biotechnology, 27(10), 2415–2423. https://doi.org/10.1007/s11274-011-0714-6
Ponraj, M., Gokila, K., & Zambare, V. (2011). Bacterial Decolorization of Textile Dye- Orange 3R. International Journal of Advanced Biotechnology and Research, 2(1), 168–177.
Puvaneswari, N., Muthukrishnan, J., & Gunasekaran, P. (2006). Toxicity assessment and microbial degradation of azo dyes. Indian Journal of Experimental Biology, 44(8), 618–626.
Rajan, S., Murugesan, A. G., Rajesh, R., & Elayarajah, B. (2013). Original Research Article Biodegradation of textile azo dyes and its bioremediation potential using seed germination efficiency. International Journal of Current Microbiology and Applied Sciences, 2(10), 496–505.
Ribeiro, A. R., & Umbuzeiro, G. de A. (2014). Effects of a textile azo dye on mortality, regeneration, and reproductive performance of the planarian, Girardia tigrina. Environmental Sciences Europe, 26(1), 22. https://doi.org/10.1186/s12302-014-0022-5
Sahasrabudhe, M. M., Saratale, R. G., Saratale, G. D., & Pathade, G. R. (2014). Decolorization and detoxification of sulfonated toxic diazo dye C.I. Direct Red 81 by *Enterococcus faecalis* YZ 66. Journal of Environmental Health Science and Engineering, 12(1), 151. https://doi.org/10.1186/s40201-014-0151-1
Santos, A., Mendes, S., Brissos, V., & Martins, L. O. (2014). New dye-decolorizing peroxidases from *Bacillus subtilis* and *Pseudomonas putida* MET94:

towards biotechnological applications. Applied Microbiology and Biotechnology, 98(5), 2053–2065. https://doi.org/10.1007/s00253-013-5041-4
Saratale, R. G., Saratale, G. D., Chang, J. S., & Govindwar, S. P. (2011). Bacterial decolorization and degradation of azo dyes: A review. Journal of the Taiwan Institute of Chemical Engineers, 42(1), 138–157. https://doi.org/10.1016/j.jtice.2010.06.006
Sathian, S., Radha, G., Shanmugapriya, V., Rajasimman, M., & Karthikeyan, C. (2013). Optimization and kinetic studies on treatment of textile dye wastewater using *Pleurotus floridanus*. Applied Water Science, 3(1), 41–48. https://doi.org/10.1007/s13201-012-0055-0
Senthilkumar, S., Perumalsamy, M., Prabhuy, H., AhmedBasha, C., & Anantharaman, N. (2012). Response surface optimization for efficient dye removal by isolated strain *Pseudomonas* sp. Open Engineering, 2(3), 425–434. https://doi.org/10.2478/s13531-012-0001-9
Senthilkumar, S., Prabhu, H. J., & Perumalsamy, M. (2013). Response surface Optimization for Biodegradation of Textile Azo dyes using isolated bacterial strain *Pseudomonas* sp. Arabian Journal for Science and Engineering, 38(9), 2279–2291. https://doi.org/10.1007/s13369-012-0507-8
Shah, M., Patel, K., & Nair, S. (2013). Microbiological removal of crystal violet dye by Bacillus subtilis ETL-2211. OA Biotechnology, 2(2), 1–7. https://doi.org/10.13172/2052-0069-2-2-548
Sharma, P., Singh, L., & Dilbaghi, N. (2009). Optimization of process variables for decolorization of Disperse Yellow 211 by *Bacillus subtilis* using Box–Behnken design. Journal of Hazardous Materials, 164(2–3), 1024–1029. https://doi.org/10.1016/j.jhazmat.2008.08.104
Singh, R. L., Singh, P. K., & Singh, R. P. (2015). Enzymatic decolorization and degradation of azo dyes – A review. International Biodeterioration & Biodegradation, 104, 21–31. https://doi.org/10.1016/j.ibiod.2015.04.027
Singh, R., Singh, R., & Singh, P. (2014). Bacterial decolorization of textile azo dye acid orange by *Staphylococcus hominis* RMLRT03. Toxicology International, 21(2), 160. https://doi.org/10.4103/0971-6580.139797
Solís, M., Solís, A., Pérez, H. I., Manjarrez, N., & Flores, M. (2012). Microbial decolouration of azo dyes: A review. Process Biochemistry, 47(12), 1723–1748. https://doi.org/10.1016/j.procbio.2012.08.014
Song, L., Shao, Y., Ning, S., & Tan, L. (2017). Performance of a newly isolated salt-tolerant yeast strain *Pichia occidentalis* G1 for degrading and detoxifying azo dyes. Bioresource Technology, 233, 21–29. https://doi.org/10.1016/j.biortech.2017.02.065
Srinivasan, V., Saravana Bhavan, P., & Krishnakumar, J. (2014). Bioremediation of Textile Dye Effluent By Bacillus and Pseudomonas spp. International Journal of Science, Environment and Technology, 3(6), 2215–2224.
Sudha, M., & Saranya, a. (2014). Microbial degradation of Azo Dyes: A review. Int. J. Curr. Microbiol. …, 3(2), 670–690.
Tahir, U., Yasmin, A., & Khan, U. H. (2016). Phytoremediation: Potential flora for synthetic dyestuff metabolism. Journal of King Saud University - Science, 28(2), 119–130. https://doi.org/10.1016/j.jksus.2015.05.009
Tan, L., He, M., Song, L., Fu, X., & Shi, S. (2016). Aerobic decolorization, degradation and detoxification of azo dyes by a newly isolated salt-tolerant yeast *Scheffersomyces spartinae* TLHS-SF1. Bioresource Technology, 203, 287–294. https://doi.org/10.1016/j.biortech.2015.12.058
Telke, A. A., Joshi, S. M., Jadhav, S. U., Tamboli, D. P., & Govindwar, S. P. (2010). Decolorization and detoxification of Congo red and textile industry effluent by an isolated bacterium Pseudomonas sp. SU-EBT. Biodegradation, 21(2), 283–296. https://doi.org/10.1007/s10532-009-9300-0
Verma, A. K., Dash, R. R., & Bhunia, P. (2012). A review on chemical coagulation/flocculation technologies for removal of colour from textile wastewaters. Journal of Environmental Management, 93(1), 154–168. https://doi.org/10.1016/j.jenvman.2011.09.012
Waghmode, T. R., Kurade, M. B., Lade, H. S., & Govindwar, S. P. (2012). Decolorization and Biodegradation of Rubine GFL by Microbial Consortium GG-BL in Sequential Aerobic/Microaerophilic Process. Applied Biochemistry and Biotechnology, 167(6), 1578–1594. https://doi.org/10.1007/s12010-012-9585-z
Witek-Krowiak, A., Chojnacka, K., Podstawczyk, D., Dawiec, A., & Pokomeda, K. (2014). Application of response surface methodology and artificial neural network methods in modelling and optimization of biosorption process. Bioresource Technology, 160, 150–160. https://doi.org/10.1016/j.biortech.2014.01.021
Yagub, M. T., Sen, T. K., Afroze, S., & Ang, H. M. (2014). Dye and its removal from aqueous solution by adsorption: A review. Advances in Colloid and Interface Science, 209, 172–184. https://doi.org/10.1016/j.cis.2014.04.002
Yan, J., Chen, D., Yang, E., Niu, J., Chen, Y., & Chagan, I. (2014). Purification and characterization of a thermotolerant laccase isoform in *Trametes trogii* strain and its potential in dye decolorization. International Biodeterioration & Biodegradation, 93, 186–194. https://doi.org/10.1016/j.ibiod.2014.06.001
Yang, Y., Wang, G., Wang, B., Li, Z., Jia, X., Zhou, Q., & Zhao, Y. (2011). Biosorption of Acid Black 172 and Congo red from aqueous solution by nonviable *Penicillium* YW 01: Kinetic study, equilibrium isotherm and artificial neural network modeling. Bioresource Technology, 102(2), 828–834. https://doi.org/10.1016/j.biortech.2010.08.125
Zhuo, R., Yuan, P., Yang, Y., Zhang, S., Ma, F., & Zhang, X. (2017). Induction of laccase by metal ions and aromatic compounds in *Pleurotus ostreatus* HAUCC 162 and decolorization of different synthetic dyes by the extracellular laccase. Biochemical Engineering Journal, 117, 62–72. https://doi.org/10.1016/j.bej.2016.09.016
Zucca, P., Cocco, G., Sollai, F., & Sanjust, E. (2016). Fungal laccases as tools for biodegradation of industrial dyes. Biocatalysis, 1(1), 82–108. https://doi.org/10.1515/boca-2015-0007