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

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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 physiochemical 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 physiochemical 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 7×10^6 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 the 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 are the key challenge (Sudha & Saranya, 2014). A feature to the complex structural varieties found in the available azo dyes, they are readily resistant to aerobic biodegradation as well as conventional wastewater treatment technologies (Huang et al., 2015). Conventional wastewater treatment technologies such as adsorption, coagulation/ flocculation, membrane filtration, electrolysis and ozonation 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 (Asud 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 (Khan 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 (Bilal et al., 2015; Lahnunhlimi & Brioshavwany, 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 yeasts) 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 (Solis 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 decolorization 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 for the removal of azo dyes as compared to the other bacterial species (Sriniwasan 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 ETI-221 in remediation of toxic crystal violet dyes, which has been employed on the optimized nutritional and environmental parameters (Shah et al., 2013). The efficacy of Bacillus sp. in remediation/ decolorisation is not only limited to anthraquinone based dyes (Acid Blue) but also highly effective towards the decolorisation of Malachite Green and Basic Blue X-GR1 (Deng et al., 2008). From several decolorization procedures, it is 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 decolorization of indigo carmine with the exploitation of Bacillus sp. MZS10 by virtue of its quinine 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 MCR from metal contaminated dyes 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 health. Natural microorganisms have been an efficient alternative to 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 interest. 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 remediating 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 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, and agitation are used as process parameters for optimization (Khan et al., 2014).

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). Illikiam 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 (Illikiam 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 underalkaline 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 bifermens 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). Ongbue and Sawidis (2011) earlier reported the optimized initial concentration of Acid Red 249 to be 100 mg/L for effective remediation by Bacillus firmus (Ongbue & Sawidis, 2011). This result was further supported by the report given by Krishna et al. (2017), where the results suggested that there was a marked decrease in the degradation efficacy when Brilliant Red X-3B, Direct Blue 6-6 and Direct Black 19 were used above 100 mg/L (Krishna 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. aeruginososa, P. putida and C. cereus attained maximized (90-94%) dye remediation from an array of synthetic dyes such as Acid Red-151, Orange II, Sulphur Black and Dimarrene Black under static condition (Rayouni 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 & Gomash, 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 remediating reactive dye, Indigo Blue, with an efficacy of 86.25% with sucrose as the sole carbon source (Ebency et al., 2013). Bheemarraddi et al. (2014) also reported that P. aeruginososa GSM3 effectively remediates 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 (Bheemarraddi 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 S. subtilis, B. cereus, B. licheniformis and Pseudomonas sp. effectively remediates 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 originalis 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. SUKI 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. SUKI was taken separately. The promising results could be attributed to the enhanced activity of laccase, veratry alcohol oxidase, azo reductase and NADH-DCIP reductase. In another report, the bioremediation of Rubine GFL using a consortium of Galactomyces yeotrichum MTCC 1360 and Brevibacillus laterosporus MTCC 2298 achieved 100% decolorization due to the activation of laccase, veratry 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 of Pseudomonas sp. and Pseudomonas sp. SUKI exhibited 99-99 % dye 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 veratry 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 FADH2 as an electron donor (Kuroda 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 (Zucza et al., 2016). These enzymes are oxygen sensitive and thus significantly inhibited by oxygen during dye 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).

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 peroxidase-dependent synthetic xenobiotics which are usually toxic and carcinogenic. However, the 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).

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 Cu2+ as the mediator. They have been studied extensively for their oxidizing effect towards various dyes (Pungare et al., 2011). Laccases oxidize the azo dye to generate a phenox radical which is subsequently re-oxidized to produce carbononium 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 because of their synergistic effect (He et al., 2015).

Table 1 Microbial enzyme mediated degradation of azo dyes.

| Enzyme                                      | Organism                        | Dye                                | Reference          |
|---------------------------------------------|---------------------------------|------------------------------------|--------------------|
| Manganese peroxidase                        | Phanerochaete sordida VK-624    | Reactive Red 120                   | Harazono et al., 2003 |
| Peroxidase, Laccase, and Azoreductase        | Exiguobacterium sp. RD3         | Reactive blue 172                  | Dhouve et al., 2008 |
| Laccase                                     | Pseudomonas sp. SU-EBT          | Congo red                          | Telke et al., 2010 |
| Laccase, Veratryl alcohol oxidase, Azoreductase and NADH-DCCP reductase | A. ochracea NCIM-1146 and Pseudomonas sp. SUK1 | Rubine GL           | H. S. Lade et al., 2012 |
| Tyrosinase, Veratryl alcohol oxidase and NADH–DCCP reductase | Brevisbacillus laterosporus | Disperse Red 54                    | Kurade et al., 2016 |
| NADH-DCCP 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-DCCP reductase, Veratryl alcohol oxidase and Tyrosinase | Brevisbacillus laterosporus | Remazol red and Rubine GL           | 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 GL        | U. U. Jadhav et al., 2009 |
| Azoreductase and NADH-DCCP reductase        | Providencia retgerri 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. greotrichum MTCC 1360 and B. laterosporus MTCC 2298 | Rubine GL | Waghmode et al., 2012 |
| Laccase                                     | Trametes troyii S0301           | Malachite blue, 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 a Time (OVAT) method is used where the independent variable is systematically 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 tools3. 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 individual 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), center points (cp), which corresponds to the middle level of the factors, and axial points (2n) (Witek-Krowiak et al., 2014). Design sp. PD1 biodegraded Congo red and indigo and two levels three-factor (2) 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 floridaus* 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 oxalidum* 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                                                     | central composite design (CCD)    | (Senthilkumar et al., 2013) |
| *Pseudomonas aeruginosa*       | Direct Blue 71                                                                  | temperature, medium pH, initial dye concentration, time | (Hafshejani et al., 2014)  |
| *Pseudomonas* sp.              | Remazol Turquoise Blue, Reactive Black 5                                       | concentrations of Dye, Carbon source, Nitrogen source | (Senthilkumar et al., 2012)  |
| *Dietzia* sp. PD1              | Congo red, Indigo carmine                                                       | pH, initial dye concentration, time | (P. Das et al., 2016)     |
| *Pseudomonas aeruginosa* PA01  | Direct Black 22                                                                | Glucose concentration, yeast extract concentration, dye concentration, inoculum size | (Mohana et al., 2008)     |
| *Stenotrophomonas maltophilia* | Reactive yellow 18, Reactive red 31, Reactive black 8, Reactive green 19        | pH, incubation time, the concentration of dye | (Kaur et al., 2015)       |
| *Proteus mirabilis*            | Reactive red 74                                                                 | Centre composite rotatable design (CCRD) |                          |
| *Cordyceps militaris* MTCC3936 | Malachite green, Bromophenol blue, Crystal violet, Acid red                     | pH, temperature                   | (Yan et al., 2014)        |
| *Pseudomonas aeruginosa* BCH   | Remazol Orange                                                                  | pH, temperature, cell mass concentration | (S. B. Jadhav et al., 2013) |
| *Pseudomonas putida* SKG-1     | Reactive orange 4                                                               | Dye concentration, sucrose, peptone, incubation time | (Garg & Tripathi, 2017)   |
| *Bacillus subtilis*            | Disperse Yellow 211                                                             | temperature, pH and initial dye concentration | ( Sharma et al., 2009)    |
| **Bacterial consortium**       | Reactive Green-19                                                               | pH, incubation temperature, Yeast extract concentration | (A. Das & Mishra, 2017)   |
| *Fomes fomentarius* laccase    | Solophenyl red 3BL                                                              | enzyme concentration, redox mediator concentration, incubation time | (Neifar et al., 2011)     |
| *Penicillium oxalidum* RF3     | Isolan Grey                                                                    | Inoculum size, media components, pH, temperature, dye concentration, incubation time | (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 Khataei et al. (2010) the three-layered feed-forward back propagation ANN model was employed to predict the decolorization efficiency of *Chaeta* 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) (Khataei 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 is also described in detail. Furthermore, 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, microorganisms-mediated remediation of azo dyes serves as an efficient, cost-effective and eco-friendly alternative to the conventional physical-chemical process for efficient removal and degradation of azo dyes from the industrial effluents.

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