

Bioefficacies of Microbes for Mitigation of Azo Dyes in Textile Industry Effluent: A review

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In recent years, India has emerged as a promising industrial hub. It has a cluster of textile, dyeing, and printing industries. The adjoining rivers/water bodies receive mostly untreated discharge from these industries. Textile industrial effluent contains various contaminants (dyes, heavy metals, toxicants, and other organic/inorganic dissolved solids) that alter the physico-chemical properties of adjoining land and waterbodies in which it is discharged, thereby degrading the water quality and subsequently affecting the landscapes in the vicinity. This ultimately affects the flora and fauna of the locale and has adverse effects on human health. Out of the total dyes (approximately 10,000 dyes) exploited in the textile dyeing and printing units, azo dyes possess a complex structure and are synthetic in origin. They contribute nearly 70% to the total effluent discharge. Biological processes are based on the ability of inhabiting indigenous microorganisms in these contaminated environments to tolerate, resist, decolorize/degrade, and mitigate the recalcitrant compounds. Exploring microbes with higher efficacy of azo dye degradation can reduce the amount of chemical discharged from the process. The present review explores the potential of microbial diversity for the development of an effective bioremediation approach. The review also includes the impact of azo dyes on the flora and fauna, as well as conventional and microbe-assisted nanoparticle technology for treatment of the textile wastewater targeting the degradation of dye contaminants.

Keywords: Bio-remediation; Azo dye; Nanoparticle; Textile effluent; Dye degradation

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INTRODUCTION

Dye pollution results *via* effluent discharge from industries such as leather, food, paper printing, pharmaceutical, textile, *etc.* (Carmen and Daniela 2012). These may amount to lethal levels, causing a variety of ecological damage under different environmental conditions. A significant amount of mainly untreated textile dye effluent (7×10^5 tons annually) is released into various waterbodies worldwide adjoining the textile dyeing and printing units, thereby changing its physico-chemical properties (APHA 1998; Hossen *et al.* 2019). Subsequently, the contaminated water takes the solvate (contaminants) to the fields in the vicinity and its consumers, adversely affecting quality of the agricultural produce, animal, and human health, causing chemosis, contact dermatitis, exophthalmose, lacrimation, permanent blindness, skin irritation, *etc.* (Sudha *et al.* 2014; Sarkar *et al.* 2017; Parmar and Shukla 2018).

Remediation of such hazardous wastes is considered to be one of the most critical environmental challenges. Compared to conventional treatment, technologies with a lack of specificity for large volumes of wastes, microorganism-based bioremediation is gaining

importance, as it has been shown to have high efficiency in mitigating, detoxifying, and degrading these contaminants (Sarkar *et al.* 2017; Tang *et al.* 2019). An understanding of the role of native microbes in complex biogeochemical reactions adds great significance for the development of microorganism-based remediation strategies. In recent years, considerable efforts have been made to explore the microbial community structure and their functional diversity in various sites contaminated with industrial effluents (Dafale *et al.* 2010; Yang *et al.* 2016). The results have indicated that these environments harbor a large number of diverse microorganisms that may have great potential for the bioremediation of these environments containing large amounts of industrial effluents.

Recently, nanotechnology utilizing nano-sized particles has become evident as a feasible alternative to the conventional methods because it is robust, readily accessible, and has a large surface area heterogeneous catalyst support (Modi *et al.* 2015; Rajput *et al.* 2017). These nano-sized particles can greatly enhance the contact between the reactants and the catalyst by increasing the exposed surface area of the active component (Cruz *et al.* 2019). Due to repeated discharge, the concentration of the dye is increasing in the environment; hence it is important to identify microbes with higher dye degradation capacity and to give effective bioremediation options for textile wastewater, such as bio-augmentation, microbial degradation, and natural attenuation to influence biostimulation either alone or with microbe-assisted nanoparticles (Ozkan *et al.* 2018). The translational efficacy has been explored to a limited extent, and microbe-assisted nanoparticles remain an enigma in environmental bioremediation at the industrial scale (Modi *et al.* 2015).

Role of Azo Dyes in Textile Dyeing Industry

Azo dyes (Acid, Disperse, Direct, Pigments, *etc.*)(namely Acid Red 183, Disperse Yellow 1/3, Disperse Orange 3/37/76, Basic Red 9, Basic Violet 14, Direct Black 38, Biebrich Scarlet, Methyl Red Sodium, tartrazine, carmoisine, *p*-di-methylaminobenzene, Sudan 1, *etc.*) constitute a major portion of the dyes used in textile industries. These are also the ones raising the biggest concern due to their mutagenic and carcinogenic nature. They link the aromatic structures with the help of one or more azo bond (-N=N-), and the cleavage of this bond biologically or chemically often releases more mutagenic and toxic end products. Azo dyes have a more intense color than anthraquinone dyes and are also relatively cheap to produce, which has resulted in their dominance in the market usability. Azo dyes form the majority of dyes being discharged into effluents. Most of the residual dyes are highly toxic by acting as a carcinogen posing a potential threat to all living organisms (Table 1).

Contaminants/Toxicants Associated with Textile Industrial Effluents

Textile industries alone discharge a wide variety of toxicants, including biodegradable organic matter, suspended solids, toxic organic compounds (phenol), synthetic dyes (such as azo, anthraquinone, phthalocyanine, and triarylmethane), heavy metal, and their conjugates. Approximately, 10 to 15% of synthetic dyes are dissipated throughout various processes in the textile dyeing and printing industry (Baban *et al.* 2003; Sudha *et al.* 2014). The pre-dominant metals associated with dyes are Pb (lead), Hg (mercury), Cr (VI) (chromium), Cd (cadmium), and As (arsenic), which are considered as highly toxic and primarily associated with textile effluents (Singh *et al.* 2017). High concentrations of these pollutants in the effluent are of solemn concern (Banat *et al.* 1996).

Table 1. Absorbance Maxima of Various Groups of Azo Dyes Facilitating Their Quantitative Indexing

S.No.	Dye Class	Category	Example and its Absorbance Maxima (nm)	Reference
1.	Acid dyes	Sodium salts of color acids that contain sulphonic acid or phenolic group	Acid Blue 40 (620 nm)	Yang <i>et al.</i> 2016
2.	Basic dyes	Basic amino group protonated under acidic condition, formation of salt linkages	Basic Fuchsin (550 nm)	Rani <i>et al.</i> 2014
3.	Direct dyes	Contains sulphonic acid group, however, these are not the point of attachment.	Direct Red 2B(530 nm)	Desai 2017
4.	Mordant dyes	Contains group that can hold metal in chelate groups or coordination complexes	Mordant Black 17 (520 nm)	Karunya <i>et al.</i> 2014
5.	Vat dyes	Like sulphur dyes, however, used in reduced form after treatment with reducing agents	Vat Red dye (540 nm)	Adebajo <i>et al.</i> 2017
6.	Reactive dyes	Forms covalent bond with fibers possessing hydroxyl or amino groups, e.g., dyes with chlorine atom	Reactive Blue 172 (570nm)	Lade <i>et al.</i> 2015
7.	Disperse dyes	Water insoluble dyes, small, and contain hydroxyl or amino group to give finite water solubility at definite temperature	Disperse Red 3B (590nm)	Tang <i>et al.</i> 2019
8.	Solvent dyes	No water solubilizing group, soluble in organic solvent	-	-

Impact of Azo Dye Contamination on Flora and Fauna

Azo dyes bio-accumulate in the environment, causing growth reduction, neurosensory damage, metabolic stress, and death of fauna and growth reduction, less productivity, and necrosis in flora. They have also been reported to be carcinogenic and mutagenic in nature. Asses *et al.* (2018) showed that the toxicity of dyes decreases after microbial treatment through phytotoxicity and micro toxicity tests. Singh *et al.* (2017) reported toxicity reduction of dyes through microbial remediation. A similar study was conducted by Lade *et al.* (2015) on the degraded metabolites of dye RB172 through acute and phytotoxicity, and the same findings were reported. Lobiuc *et al.* (2018) assessed CR toxicity towards *Lemna minor* and reported reduction in root growth, total frond surface and fresh mass reduced from 5 ppm dye concentration, whereas above 2500 ppm concentration, complete plant growth was inhibited. Khandare *et al.* (2013) demonstrated the metabolism fate of Direct Red 5B by *P. grandiflora*, *P. putida*, and their consortium with the help of GC–MS analysis. Gita *et al.* (2018) reported that the specific growth rate decreased with the increase in concentration of Optilan Red; maximum percentage inhibition was 66.6% and 79.4% for total chlorophyll and carotenoid, respectively (at 50 ppm). Khatun (2017) revealed severe histopathological effects of silk dye waste effluent on the tissues of both intestines and stomachs of Swiss albino mice. Atrophy of musculature, degeneration of mucosal epithelial cells characterized by nuclear pyknosis, cytoplasmic vacuolization, and nuclear fragmentation were reported along with damage in the Brunner's gland and the crypts of Lieberkuhn. A study performed on mung bean seed germination demonstrated reduction in radicle-plumule growth and percent seed

germination (Khan and Malik 2017). Desai (2017) demonstrated phytotoxicity studies on mung seeds and reported that good germination and shoot, root length of the plants were observed for degraded dye metabolite exposed seeds after comparing with the control using *Klebsiella* sp. and *Staphylococcus* sp. Laxmi and Nikam (2015) demonstrated toxicity reduction of the metabolites formed after dye degradation, as the growth of seedlings and seed germination percentage of *Guizotia abyssinica* were at par with water and the decolorized dye sample (using *A. flavus*). Rani *et al.* (2014) reported that seeds of *Triticum aestivum* inoculated with textile dye solution of Malachite Green treated with *A. niger* and *P. chrysosporium* show germination, while uninoculated solution hindered germination. Rajeswari *et al.* (2014) used *Lysinibacillus sphaericus* and *Stenotrophomonas maltophilia* treated solutions on *Triticum aestivum* and the human embryonic kidney cell line (HEK 293) for evaluation of phytotoxicity and cytotoxicity.

Another study illustrated toxicity reduction using a phytotoxicity assay on the seeds of *Phaseolus mungo* and *Sorghum vulgare*, as the seeds were more sensitive towards the dye in comparison to its by-product (Kalyani *et al.* 2008). Sharma *et al.* (2007) conducted serum biochemical and haematological studies on Swiss albino rats and stated that the values of white blood cells (WBC), red blood cells (RBC), packed cell volume (PCV), haemoglobin (Hb), and mean corpuscular hemoglobin concentration (MCHC) significantly decreased in wastewater-exposed animals (12 to 46%) with respect to control animals (potable water). Further, reduction in RBC size (13 to 27%) and the shape modification (poikilocytosis) was observed. The serum biochemical parameters alanine transaminase (ALT), aspartate aminotransferase (AST), creatinine, urea, and bilirubin significantly increased (5 to 97%), while cholesterol, glucose, total protein, albumin, and globulin contents decreased (8 to 53%). Sponza and Isik (2004) published that *Daphnia magna* tests and anaerobic toxicity assays (ATA) respiration/inhibition showed reduction in toxicity of C.I Direct Red 28. The LC50 of dyes revealed toxicity of blue>yellow>red>orange dye (Sani *et al.* 2018). Another study reported that the Procion Red MX-5B dye solution after treatment with *A. niger* showed an increase in toxicity, thereby retarding the growth of *Lactuca sativa* seeds by 43% and mortality to 100% in *A. salina* larvae (Almeida and Corso 2014). Zhang *et al.* (2012) evaluated the toxicity of effluent samples from sewage treatment plants (STPs) using bioassays with zebrafish, which indicated high acute toxicity and genotoxicity. Przystaś *et al.* (2012) conducted zootoxicity and phytotoxicity tests with *Daphnia magna* and *Lemna minor*, respectively, of degraded by-products. The degradation of brilliant green correlated with the decrease of zootoxicity (*D. magna*) and phytotoxicity (*L. minor*) (Table 2).

Table 2. Toxicity Analysis of Azo Dyes on Flora and Fauna

S.No.	Dye	Plant/Animal	Impact	Reference
1.	Congo Red	Plant (<i>Zea mays</i> and <i>Solanum lycopersicum</i>)	Reduced significantly the germination rate, shoot, and root length	Asses <i>et al.</i> 2018
2.	Textile effluent	Plant (<i>Triticum aestivum</i> , <i>Phaseolus mungo</i> , and <i>Vigna radiata</i>)	Number of germination and length of radical and plumule were less in untreated effluent	Singh <i>et al.</i> 2017
3.	Reactive Blue 172	Animal (<i>Daphnia magna</i>); Plant (<i>Sorghum vulgare</i> and <i>Phaseolus mungo</i>)	Acute test showed 100% mortality of <i>D. magna</i> in untreated dye RB 172;	Lade <i>et al.</i> 2015

			Number that germinated reduced 60 to 70% and length of radical and plumule reduced	
4.	Procion Red MX-5B	Animal (<i>Artemia salina</i>); Plant (<i>Lactuca sativa</i>)	No toxicity on <i>A. salina</i> . Inhibition of root growth on <i>L. sativa</i>	Almeida and Corso 2014
5.	Direct Red 28	Plant (<i>Lemna minor</i>)	Reduction in total frond surface and root growth, dye accumulation in tissues, necrosis, chlorophyll a contents decreased; significant inhibition of PSII efficiency, chromosomal aberrations significantly increased and fresh mass reductions at different dye concentration from 5 to 5000 ppm; complete growth inhibition at 2500 ppm	Lobiuc <i>et al.</i> 2018
6.	Direct Red 5B	Plant (<i>Sorghum vulgare</i> and <i>Phaseolus mungo</i>)	Inhibitory effect on shoots and root length of plants; Low percentages of germination of <i>P. mungo</i> and <i>S. vulgare</i> with reduced lengths of the plumule and radical	Khandare <i>et al.</i> 2013
7.	Optilan Red	Microalgae (<i>Chlorella vulgaris</i>)	Specific growth rate decreased at 20 ppm, EC50 23.16ppm, protein reduced, 66.6% and 79.4% for total chlorophyll and carotenoid, respectively, and elemental composition reduced at 50 ppm	Gita <i>et al.</i> 2018
8.	Silk dye waste effluent	Animal (Swiss albino mice)	Histopathological effects in tissues of stomach, intestine, atrophy of musculature, degeneration of mucosal epithelial cells characterized by nuclear pyknosis, cytoplasmic vacuolization and nuclear fragmentation were reported along with damage in Brunner's gland and crypts of Lieberkuhn.	Khatun 2017
9.	Textile effluent	Plant (<i>Vigna radiata</i>)	Reduction in percent seed germination and radicle-plumule growth	Khan and Malik 2017
10.	Dark Red 2B	Plant (<i>Phaseolus mungo</i>)	Reduction in germination and shoot, root length	Desai 2017
11.	Reactive Navy Blue M3R, Reactive Red M8B, Reactive	Plant (<i>Guizotia abyssinica</i>)	Percent germination, plumule, radical, protein, and total carbohydrate content reduction	Laxmi and Nikam 2015

	Green HE4B, Reactive Orange M2R, Reactive RedM5B, Direct Orange RS, Direct Black BT, Direct Blue GLL, and Direct Sky Blue FF			
12.	Malachite Green	Plant (<i>Triticum aestivum</i>)	Dye inhibited the germination of wheat seeds	Rani <i>et al.</i> 2014
13.	Yellow ME4GL, Blue RR, Red RR, Yellow RR, Red M5B, Blue MR, Deep Black RR, Yellow MERL, Red ME4BL, and Golden Yellow MR (mixture)	Plant (<i>Triticum aestivum</i>); Animal (human embryonic kidney cell line (HEK 293))	Germination percent, length of plumule and radical reduction. Dye solution interfere with the colour, also the cells were not clearly detected when observed under inverted microscope	Rajeswari <i>et al.</i> 2014
14.	Reactive Red BL1	Plant (<i>Sorghum vulgare</i> and <i>Phaseolus mungo</i>)	Reductions in germination percent, and in length of plumule and radical	Kalyani <i>et al.</i> 2008
15.	Textile effluent	Animal (Swiss albino rats)	Values of WBC, RBC, PCV, Hb, and MCHC significantly decreased, reduction in RBC size (13 to 27%), and the shape modification (poikilocytosis). The serum biochemical parameters ALT, AST, creatinine, urea, and bilirubin significantly increased (5 to 97%), while cholesterol, glucose, total protein, albumin, and globulin contents decreased (8 to 53%)	Sharma <i>et al.</i> 2007
16.	Direct Red 28	Animal (<i>Daphnia magna</i>)	Increase in mortality rate of the test organism	Sponza and Isik 2004
17.	Textile dye (defined by colours, blue, red, orange, and yellow)	Animal (<i>Artemia salina</i>)	LC50 of the various dyes on the test animals indicated that the blue dye was the most toxic, followed by yellow, red, and orange.	Sani <i>et al.</i> 2018
18.	Textile effluent	Animal (<i>Danio rerio</i>)	High acute toxicity and genotoxicity	Zhang <i>et al.</i> 2012
19.	Brilliant green and Evans blue	Animal (<i>Daphnia magna</i>) and plant (<i>Lemna minor</i>)	Decolourization of brilliant green was connected with decrease of zootoxicity (<i>D. magna</i>) and phytotoxicity (<i>L. minor</i>). Removal of Evans blue was connected with no changes in zootoxicity and decrease of phytotoxicity in most of samples.	Przystaś <i>et al.</i> 2012

STATUS OF TEXTILE EFFLUENT AND MICROBIAL AZO DYE DEGRADATION IN INDIA AND THE WORLD

Various studies have been conducted worldwide on effluent discharge, its characteristics, and treatment. Hasan and Miah (2014) investigated the impact of textile mill effluent on surface water and reported that concentrations of electrical conductivity (EC), biological oxygen demand (BOD), total dissolved solids (TDS), Na^+ , Cl^- , NH_4^+ , NO_3^- , HCO_3^- , SO_4^{2-} , PO_4^{3-} , and toxic metals (Cd, Cr, and Pb) of the collected effluent samples exceeded the standard levels and were unsuitable for drinking, domestic purposes, or irrigation purposes. Starovoitova and Odido (2014) reported that the compounds categorized as carcinogenic to human beings were used in the industry as metal/complex/chrome/mordants dye. However, in India, it has been reported that the textile effluents collected from Sanganer (Jaipur) had higher temperature, pH, EC, total suspended solids (TSS), total dissolved solids (TDS), chloride content, and hardness compared to the limits prescribed by World Health Organization (WHO) guidelines for textile industrial effluent (Sharma *et al.* 2013; Jaishree and Khan 2014; Rahi *et al.* 2018). Satija and Bhatnagar (2017) published that the wastewater collected from dyeing and printing industries revealed slightly alkaline pH (7.7 to 13.02); and significant TDS (3337.25 to 1494.6 $\mu\text{S}/\text{cm}$); TSS (22.20 to 5.8 NTU); and cations and anions (Ca^{+2} : 427.6 to 175, Mg^{+2} : 174.4 to 77.8, Cl^- : 2028 to 1039, F^- : 16.8 to 9.2, SO_4^{-2} : 304.6 to 182.8, CO_3^{-2} : 144.2 to 53.6, and HCO_3^{-2} : 408.8 to 180.2) that was higher than the desired limits. Another study evaluated physico-chemical parameters, such as pH, color, total hardness, COD, BOD, TSS, TDS, turbidity, chlorides, sulphides, silica, calcium, iron, oil, and grease, of the effluent and confirmed that all the parameters studied were above guideline permissible limits, excluding calcium, sulphide, and iron, in the context of the water quality standards of the Bureau of Indian Standard (BIS), Central Pollution Control Board (CPCB), and National Environment Quality Standards (NEQS) (Mahawar and Akhtar 2015; Patel *et al.* 2015; Sriram and Reetha 2015; Elango *et al.* 2017).

Various microorganisms have been explored throughout these years for azo dye degradation, namely bacteria, fungi, and algae. Ajaz *et al.* (2019) reported that *Alcaligenes aquatilis* decolorizes 82% of Synazol red 6HBN in 4 days at 37 °C, pH 7, under static conditions with yeast extract and saw dust as nitrogen and carbon sources. Another study confirmed that indigenous *Bacillus cereus* AZ27, *Alcaligenes faecalis* AZ26, and *Bacillus* sp. AZ28 have the potential to mitigate various dyes with more than 25% of degradation optimized using Novacron Super Black G (NSB-G) (Hossen *et al.* 2019). Another study illustrated that the bacterial consortium comprising of *Sphingomonas paucimobilis*, *Rhizobium radiobacter*, and *Bacillus subtilis* removes heavy metals from industrial wastewater and decolorizes Methyl Orange (MO) and CR textile azo dyes better than their corresponding single cultures, correlating to synergistic activity of different metabolites of bacterial cultures and protection of cells from toxic pollutants that was provided by Calcium alginate matrices (Allam 2017). Lade *et al.* (2015) used single bacterium *Providencia rettgeri* strain HSL1 that completely decolorized 50 mgL^{-1} of dye C.I. Reactive Blue 172 (RB172) in 20 h at 30 ± 0.2 °C under microaerophilic conditions and showed considerable reduction in total organic carbon (TOC) (52%) and chemical oxygen demand (85%) contents, which correlated with nicotinamide adenine dinucleotide-dichlorophenol indophenols (NADH-DCIP) reductase (88%) and azoreductase (159%) activity. Singh *et al.* (2017) demonstrated that bacterial strains of *Enterobacter asburiae* and *E. cloacae*, used as consortium, efficiently decolorized (up to 98%) at pH 1.67, 32°C within 10 min

under aerobic condition. Khan and Malik (2018) reported that *Arthrobacter soli* BS5 degrades textile dye reactive black 5 with maximum degradation of 98% at pH 5 to 9, 37°C after 120 h of incubation. Another study published that *Klebsiella* sp. and *Staphylococcus* sp. decolorizes 200 ppm of Direct Red 2B dye by 98.8% and 98.7%, respectively (Desai 2017). Lalnunhlmi and Krishnaswamy (2016) demonstrated that bacterial consortium isolated from soil samples of a saline environment decolorizes 200 mg/L of Direct Blue 151 (DB151) and Direct Red 31 (DR 31) by 97.6% and 95.2%, respectively, within 5 days, which further improved supplementation of sucrose and yeast extract that were used as carbon and nitrogen sources. Vimala *et al.* (2015) isolated bacteria as *Pseudomonas* sp., *Citrobacter* sp., *Escherichia coli*, and *Micrococcus* sp. and reported that plasmid was present in all isolates. *Micrococcus* sp. was reported to work well in an adapted environment, while *E. coli* showed the same decolorization potential in adapted and non-adapted environments. Singh *et al.* (2014) identified *Staphylococcus hominis* as a potential degrader of Acid Orange dye up to 600 mg/L at pH 7.0, 35°C, after 60 h of incubation with yeast extract and glucose supplementation (Table 3). Khandare *et al.* (2013) utilized bacterial and plants consortium of *Pseudomonas putida* and *Portulaca grandiflora* and reported complete degradation of a sulfonated diazo dye. Enzymes involved were reported as 2,6-dichlorophenol indophenol reductase, riboflavin reductase, lignin peroxidase, and tyrosinase in *P. grandiflora* and veratryl alcohol oxidase, laccase, and 2,6-dichlorophenol indophenol reductase in *P. putida*.

Table 3. Degradation of Various Dyes using Bacteria

#	Dye	Bacteria	Mechanism	Reference
1	Synazol Red 6HBN	<i>Alcaligenes aquatilis</i>	Mineral salt medium, 4 days, 37 °C, pH 7, static with yeast extract, and saw dust	Ajaz <i>et al.</i> 2019
2	Novacron Super Black G	<i>Bacillus cereus</i> AZ27, <i>Alcaligenes faecalis</i> AZ26, and <i>Bacillus</i> sp. AZ28	Nutrient broth medium, 4 days, 37 °C, pH 8, and at static condition	Hossen <i>et al.</i> 2019
3	Congo Red, Phenol Red, Direct Yellow, Direct Red, Acid Orange, Direct Violet, Direct Blue, Direct Pink	<i>Staphylococcus</i> sp., <i>Bacillus</i> sp., <i>Pseudomonas</i> sp., <i>Enterobacter</i> sp., <i>Micrococcus</i> sp., and <i>Klebsiella</i> sp.	Nutrient broth medium, 3 days	Islam <i>et al.</i> 2017
4	Methyl Orange and Congo Red	<i>Sphingomonas paucimobilis</i> , <i>Rhizobium radiobacter</i> , and <i>Bacillus subtilis</i> (Consortia)	Nutrient broth medium, 4 days, 37 °C, pH 7.5, shaken at 120 rpm (free cell vs. immobilized consortia)	Allam 2017

5	Red and Green dye	<i>Bacillus</i> sp., <i>Pseudomonas</i> sp., and <i>Aspergillus</i> sp.	Nutrient broth 1 day for bacteria and Sabouraud's Dextrose broth 3 days for fungi, 37 °C, occasional shaking	Fatima and Alamgir 2015
6	Reactive Blue 172	<i>Providencia rettgeri</i> strain HSL1	Wheat bran medium, 20 h, 30±0.2°C, pH 7, shaken at 120 rpm, under microaerophilic conditions (correlated with NADH-DCIP reductase and azoreductase activity)	Lade <i>et al.</i> 2015
7	Textile effluent	<i>Enterobacter asburiae</i> and <i>Enterobacter cloacae</i> (consortium)	Nutrient broth, 10 min, 32 °C, pH 1.67, under aerobic conditions	Singh <i>et al.</i> 2017
8	Reactive Black 5	<i>Arthrobacter soli</i> BS5	Nutrient broth, 5 days, 37 °C, pH 5 to 9, static	Khan and Malik 2018
9	Direct Red 2B	<i>Klebsiella</i> sp. and <i>Staphylococcus</i> sp.	Bushnell Hass minimal salt medium, 5 days, 37°C, pH 7, rotary shaker at 100 rpm, supplementation 1% glucose	Desai 2017
10	Direct Blue 151 and Direct Red 31	<i>Bacillus flexus</i> strain NBN2, <i>Bacillus cereus</i> strain AGP-03, <i>Bacillus cytotoxicus</i> NVH 391-98, and <i>Bacillus</i> sp.L10	Mineral salt medium, 5 days, 36 °C, pH 9.5, on shaker, supplementation of sucrose and yeast extract	Lalnunhlmi and Krishnaswamy 2016
11	Procine and Direct dyes	<i>Pseudomonas</i> sp., <i>Citrobacter</i> sp., <i>Escherichia coli</i> , and <i>Micrococcus</i> sp.	Nutrient broth, 1 day, 37 °C, pH 7, rotary shaker	Vimala <i>et al.</i> 2015
12	Acid Orange	<i>Staphylococcus hominis</i>	Bushnell and Haas medium, 60 h, 35 °C, pH 7, static supplemented with glucose and yeast extract	Singh <i>et al.</i> 2014

Al-Tohamy *et al.* (2020) illustrated that the decolorizing ability of yeast *Sterigmatomyces halophilus* on Reactive Black 5 depends on NADH-dichlorophenol indophenol (NADH-DCIP) reductase and lignin peroxidase (LiP). Asses *et al.* (2018) used *Aspergillus niger* for biodegradation of Congo red (CR), an azo dye. The decolorization rate reached 97% on inoculation of 2 g mycelia and 200 mg/L of dye in 6 days at pH 5, 28 °C, and 120 to 150 rpm, which correlated with manganese peroxidase and lignin peroxidase production. A study reported *Pseudomonas* sp. as having high efficacy in the mitigation of azo dyes and *Pseudomonas* sp., *Micrococcus* sp., and *Bacillus* sp. in removing heavy metals ranging up to 350 to 550 µg/mL (Islam *et al.* 2017). Cheng *et al.* (2016) screened white-rot fungi for their azo dyes degradation capacity using Biebrich Scarlet (C.I. 26905), Direct Blue 71 (C.I. 34140), Orange G (C.I. 16230), and Ponceau 2R (C.I. 16450). *Coriolopsis* sp. strain arf5 was identified as a microbe that completely degraded all four

dyes in the shortest time interval when supplemented with an additional carbon source (glucose) and nitrogen-limiting conditions. At the same time, Yang *et al.* (2016) isolated freshwater fungal strains from submerged woods and used them for the degradation of seven synthetic dyes. Another study used indigenous bacterial and fungal isolate for degradation of red and green textile dyes at 10, 50, 100, and 150 mg/L. *Pseudomonas* sp. demonstrated higher dye decolorization (67% for red and 73% for green dye) in comparison to *Aspergillus* sp. (59% for red dye and 70% for green dye) in three days (Fatima and Alamgir 2015). Almeida and Corso (2014) reported that *Aspergillus terreus* and *A. niger* decolorizes 30% of Procion Red MX-5B dye in 3 h of biosorption (100%). The ultraviolet (UV)–visible (VIS) spectroscopy analysis indicated the removal of the dye molecules occurred without statistically significant molecular changes. Laxmi and Nikam (2015) isolated *A. flavus*, *A. niger*, *Fusarium oxysporium*, and *Penicillium notatum* and used them for the decolorization of azo textile dyes. The study reported that *A. niger* decolorized basic fuchsin (81.85%)>Nigrosin (77.47%)>Malachite green (72.77%) >dye mixture (33.08%), while *Phanerochaete chrysosporium* decolorized Nigrosin (90.15%)>basic fuchsin (89.8%)>Malachite green (83.25%)>mixture (78.4%) under shaking condition (Rani *et al.* 2014) (Table 4).

Table 4. Degradation of Various Dyes using Fungi

#	Dye	Microbe	Mechanism	Reference
1	Reactive Black 5, Reactive Red 120, Reactive Blue 19, Acid Scarlet GR, and Azure B	<i>Sterigmatomyces halophilus</i>	Minimal Salt (MS) medium, 24 hours, 115 °C, pH 5, 50 mgL ⁻¹ dye concentration, supplemented with carbon and energy sources, including glucose, ammonium sulfate, and yeast extract	Al-Tohamy <i>et al.</i> 2020
2	Congo Red	<i>Aspergillus niger</i>	Synthetic nutrient broth medium, 6 days, 28 °C, pH 5, shaken at 120 to 150 rpm (correlated with manganese peroxidase and lignin peroxidase)	Asses <i>et al.</i> 2018
3	Biebrich Scarlet, Direct Blue 71, Orange G, and Ponceau 2R	<i>Corioloopsis</i> sp. Strain arf5	Kirk's basal medium, 7 to 12 days, room temperature to 35 °C, pH 4.5 to 6, static under nitrogen-limiting conditions and an additional carbon source (glucose)	Cheng <i>et al.</i> 2016
4	Acid Blue 40, Acid Blue 193, Acid Blue 62, Acid Blue 113, Acid Red 73 Reactive Red 11, Reactive Blue 74	<i>Myrothecium verrucaria</i> , <i>Colletotrichum dematium</i> , <i>Corynespora cassicola</i> , <i>Dictyosporium zhejiangensis</i> , <i>Plectosporium tabacinum</i> , <i>Fusarium thapsinum</i> , <i>Alternaria alternate</i> , <i>Acrogenospora sphaerocephala</i> , and <i>Ceriporia lacerate</i>	Malt agar medium, 7 days, 25 °C	Yang <i>et al.</i> 2016

5	Procion Red MX-5B	<i>Aspergillus terreus</i> and <i>Aspergillus niger</i>	Aqueous solution, 3 h (biosorption), 1 day to 14 days (biodegradation), 30±1 °C, and pH 4	Almeida and Corso 2014
6	Reactive (Navy Blue M3R, Red M8B, Green HE4B, Orange M2R, RedM5B), Direct (Orange RS, Black BT, Blue GLL, Sky Blue FF)	<i>Aspergillus flavus</i> , <i>Aspergillus niger</i> , <i>Fusarium oxysporium</i> , and <i>Penicillium notatum</i>	Potato dextrose broth, 3 to 7 days, 30 °C, pH 5, in rotary shaker at 120 rpm, biotransformation enzymes (lignin peroxidase>laccase>manganese peroxidase>tyrosinase)	Laxmi and Nikam 2015
7	Malachite Green, Nigrosin Disodium, and Basic Fuchsin	<i>Aspergillus niger</i> , and <i>Phanerochaete chrysosporium</i>	Potato dextrose broth, 10 days, 25 °C, pH 7, rotary shaker at 100 rpm	Rani <i>et al.</i> 2014

Ishchi and Sibi (2020) illustrated that microalgae *Chlorella vulgaris* have azo dye degrading capacity using Reactive Black 5, Direct Blue 71, and Disperse Red 1. The decolorizing results were shown to depend on initial dye concentration and different pH for different dyes. Tang *et al.* (2019) reported that Disperse Red 3B decolonization occurred better in the consortium of *Chlorella sorokiniana* XJK and *Aspergillus* sp. XJ-2(98.09%) with respect to their single system, while the removal rate of TP (total phosphorus) 83.9%, COD (chemical oxygen demand) 93.9%, and ammonia nitrogen 87.6% under the optimized conditions were achieved due to lignin peroxidase and manganese peroxidase enzyme activities. Hernandez-Zamora *et al.* (2015) showed that through biodegradation and biosorption processes, *Chlorella vulgaris* removed 83 and 58% of congo red dye at concentrations of 5 and 25 mg L⁻¹, respectively. Khataee *et al.* (2010) used Xanthophyta alga, *Vaucheria* sp. to degrade Malachite green and concluded that degradation is inversely proportional to initial dye concentration and directly proportional to pH, temperature, and algal biomass. El-Sheekh *et al.* (2009) investigated the decolorizing potential of *Elkatotrix viridis*, *Chlorella vulgaris*, *Nostoc linckia*, *Lyngbya lagerlerimi*, *Oscillatoria rubescens*, and *Volvox aureus* using methyl red, orange II, basic cationic, G-Red (FN-3G), and basic fuchsin and concluded that decolorizing using *C. vulgaris* or *N. Linckia* with G-Red or methyl red, respectively, induced the algal azo dye reductase enzyme by 72 and 71% at the same order (Table 5).

Table 5. Degradation of Various Dyes using Algae

#	Dye	Microbe	Mechanism	Reference
1	Disperse Red 3B	<i>Chlorella sorokiniana</i> XJK and <i>Aspergillus</i> sp. XJ-2 (consortia)	BG11 and Czapek's medium, respectively, 4 days, 30 °C, pH 7, and shaken at 170 rpm (due to lignin peroxidase and manganese peroxidase enzyme activities)	Tang <i>et al.</i> 2019
2	Congo red	<i>Chlorella vulgaris</i>	Bold's basal mineral medium, 25±3 °C, 3 days	Hernandez-Zamora <i>et al.</i> 2015
3	Malachite green	Xanthophyta alga, <i>Vaucheria</i> sp.	Dye solution, pH 1.5 – 8.5, temperature 5- 45 °C and initial dye concentration 2.5 – 17.5 mg L ⁻¹	Khataee <i>et al.</i> 2010
4	Methyl red, orange II, basic cationic, G-Red (FN-3G) and basic fuchsin	<i>Elkatothrix viridis</i> , <i>Chlorella vulgaris</i> , <i>Nostoc linckia</i> , <i>Lyngbya lagerlerimi</i> , <i>Oscillatoria rubescens</i> , and <i>Volvox aureus</i>	Bold's basal mineral medium, pH 7, 25±1 °C, continuous light intensity 5000 to 3000 lux on rotary shaker	El-Sheekh <i>et al.</i> 2009
5	Reactive Black 5, Direct Blue 71 and Disperse Red 1.	<i>Chlorella vulgaris</i>	Mineral salt media, 100 mg L ⁻¹ dye, 12 days, 30 °C, dye concentration 100-500 mg L ⁻¹ in static conditions	Ishchi and Sibi 2020

Ajaz *et al.* (2019) used high-performance liquid chromatography (HPLC), thin layer chromatography (TLC), gas chromatography–mass spectroscopy (GC-MS), and Fourier transform infrared spectroscopy (FTIR) analysis to confirm the cleavage of azo bond. The bacterially treated FTIR sample showed an absence of peaks at the 1532 cm⁻¹ and 1612 cm⁻¹ wavelengths, demonstrating the breakdown of the azo bond. Ayed *et al.* (2011), Ahmed *et al.* (2016), and Hossen *et al.* (2019) investigated dye decolorization using UV-VIS spectrophotometry and FTIR analysis. The statistically significant difference in UV-VIS absorbance spectra and the FTIR spectrum of the decolorized dye from those of the parent dye revealed that the dye was mitigated by the bacterial isolates. Tang *et al.* (2019) reported that UV-VIS spectrophotometry, FTIR, and GC-MS analysis revealed that the colored functional groups of Dispersed Red 3B were broken down into less toxic small molecular compounds. Asses *et al.* (2018) characterized degraded metabolites using liquid chromatography – tandem mass spectrometry (LC-MS/MS) of Congo Red mainly by

peroxidases activities. Yang *et al.* (2016) reported that biotransformation occurred after fungal biodegradation of synthetic dyes as it formed new absorbance peaks (Table 6).

Khandare *et al.* (2013) and Singh *et al.* (2017) used UV-VIS and FTIR analysis for dye degradation analysis. Laxmi and Nikam (2015) confirmed that dye decolorization occurred through degradation using UV-VIS spectrophotometric and high-performance thin layer chromatography (HPTLC) analysis.

Table 6. Degradation Analysis of Dyes Using High Throughput Spectral Scan

#	Dye	Analysis	Scan Interpretation	Reference
1.	Synazol Red 6HBN	HPLC, thin layer chromatography (TLC), GC-MS, and FTIR	Cleavage of azo bond. The chromatogram of the untreated dye sample showed three detectable peaks at retention times of 1.80, 2.88, and 5.00 min while treated dye sample showed peaks at retention times of 1.99, 2.30, 2.95, and 3.76 min. TLC showed two bands in treated with reference to one band of control. GC-MS analysis confirmed that azo dye enzymatically converted into various end products. The absence of peaks of wavelength 1612 cm^{-1} and 1532 cm^{-1} in bacterially treated FTIR sample demonstrated the cleavage of azo bond.	Ajaz <i>et al.</i> 2019
2.	Methyl Red, Novacron Super Black G	UV-VIS spectrophotometry and FTIR analysis	Biodegradation of parent dye Methyl Red peak at 435 nm decreased without any shift in λ_{max} . Novacron Super Black G λ_{max} shifted from 600 to 410, 378, and 373nm by different microbes. The shift of peaks of wavelength from 1604, 1384, 1124, and 535 cm^{-1} to 1603, 1383, 1114, and 530 cm^{-1} in bacterially treated FTIR sample of MR, while in Novacron Super Black G from 1134, 1051, 1004, 1342, 1639, and 632 to 873, 931, 1294, 1384, 1429, 1581, 881, 943, 1174, 1355, 1382, 1477, 1652 or 885, 889, 1020, 1112, 1247, 1344, 1382, 1479, and 1529 by different bacteria, respectively	Ayed <i>et al.</i> 2011 and Hossen <i>et al.</i> 2019
3.	Disperse Red 3B	UV-VIS spectrophotometry, FTIR, and GC-MS analysis	Parent dye broken down into less toxic small molecular compounds. Dye peak at 590 nm decreased until it disappeared without any shift in λ_{max} . The absence of peaks at 797.3 cm^{-1} in bacterially treated FTIR sample demonstrated the	Tang <i>et al.</i> 2019

			cleavage of aromatic ring structure. GC-MS confirmed enzymatic degradation of dye.	
4.	Congo Red	Liquid chromatography – tandem mass spectrometry (LC-MS/MS)	LC-MS/MS confirmed degraded metabolites through peroxidases activities.	Asses <i>et al.</i> 2018
5.	Acid Blue 40, Reactive Red 11, Acid Blue 193, Acid Blue 62, Acid Blue 113, Reactive Blue 74, and Acid Red 73	UV-VIS spectrophotometry	Formation of new peak confirming biotransformation occurred after fungal biodegradation of synthetic dyes.	Yang <i>et al.</i> 2016
6.	Textile effluent	UV-VIS analysis and FTIR analysis	Almost complete adsorption or degradation of dye. Textile effluent peak at 490 and 650 nm completely disappeared without any shift in λ_{max} . The absence of peaks of wavelength 3445, 1636.7, 1123.63, 1096, and 599.76 cm^{-1} and formation of new peaks in bacterially treated FTIR sample demonstrated the cleavage of azo bond.	Singh <i>et al.</i> 2017
7.	Direct Red 5B	HPLC, GC-MS, and FTIR analysis	Biotransformation of the dye into different metabolites. The chromatogram of the untreated dye sample showed three detectable peaks at retention time of 1.77 min, while treated dye sample showed peaks at retention time of 3.02, 3.38, 3.76, and 7.45 min. GC-MS indicates the action of enzymes on the complex structure of dye molecule metabolized into simpler chemical species. FTIR peaks at 1754.7, 1619.3, 1546.7, 1486, 1286.8, 1134.7, and 1045.1 cm^{-1} shifted indicating presence of sulfo groups in the products but a decrease in their number indicates more deamination and desulfonation of DR5B.	Khandare <i>et al.</i> 2013
8.	Reactive Navy Blue M3R, Reactive Red M8B, Reactive Green HE4B, Reactive Orange M2R, Reactive	UV-visible spectrophotometric and HPTLC analysis	Biotransformation of the dye into different metabolites. HPTLC chromatogram showed the absence of control dye band in the treated sample metabolites lane, which indicates their complete transformation.	Laxmi and Nikam 2015

	RedM5B, Direct Orange RS, Direct Black BT, Direct Blue GLL, and Direct Sky Blue FF			
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TREATMENT APPROACHES

Conventional Treatment (Primary, Secondary, Tertiary, and Pain Point After Tertiary Treatment as per CPCB Guidelines)

Textile industry is one of the largest industries in the world, and different fibers, such as cotton, silk, and wool, as well as synthetic fibers, are all pre-treated, colored, and after-treated using a large amount of water and other chemicals. The pollutants include various dyes, starches, and detergents that undergo various physio-chemical changes that consume dissolved oxygen from the receiving stream and destroy aquatic life. Such organics should be removed to prevent septic conditions and avoid rendering the water stream unsuitable for municipal, industrial, agricultural, and residential uses. Treatment of wastewater reduces the waste, prevents negative effects, and makes positive effects on its further usage. Effluent treatment plants (ETPs) treat the water that comes out of these industries. Parameters including pH, color, biological oxygen demand, chemical oxygen demand, oil and grease, total dissolved solids, total suspended solids, *etc.* are evaluated in compliance with the Central Pollution Control Board (CPCB). Preliminary treatment level comprises of physical separation of large-sized impurities like plastics, polythene bags, paper, wooden logs, *etc.* This is done either through clarification that uses a belt to remove large-sized impurities or sedimentation that uses gravity for the separation.

Further treatment of the effluent is characterized into the following categories: Primary, Secondary, and Tertiary. Primary treatment is a physio-chemical method used to remove suspended solids and treat parameters such as pH, oil, and grease using coagulation, chemical precipitation, and oxidation. Sodium hydroxide, sodium carbonate, and calcium carbonate are used to treat the pH of acidic effluent, while sulphuric acid or hydrochloric acid are used to treat the pH of alkaline effluent. Alum ($Al_2(SO_4)$) is used as a chemical coagulant, further, a chemical flocculent is added to aid precipitation by bringing fine particles together to form large masses.

Secondary treatment involves biological treatment of the effluent to remove organic and inorganic impurities using microbes, *i.e.* bacteria or fungi. Mainly aerobic treatment is performed, *i.e.* in the presence of oxygen. Nitrifying bacteria are used to convert the compounds into other by-products.

Tertiary treatment finally processes the water to meet the disposable guidelines for further reusing, recycling, or disposing into the environment. It removes the remaining impurities such as inorganic compounds, bacteria, parasites, *etc.* Alum is further added to remove any additional particle by grouping them so that they are being removed at the last stage. Chlorine is added to disinfect the treated wastewater from bacteria, fungi, parasites, *etc.* Sodium bisulphate is then added to remove excessive chlorine. The wastewater is then centrifuged before being discharged through the outlet into the environment (Fig.1).

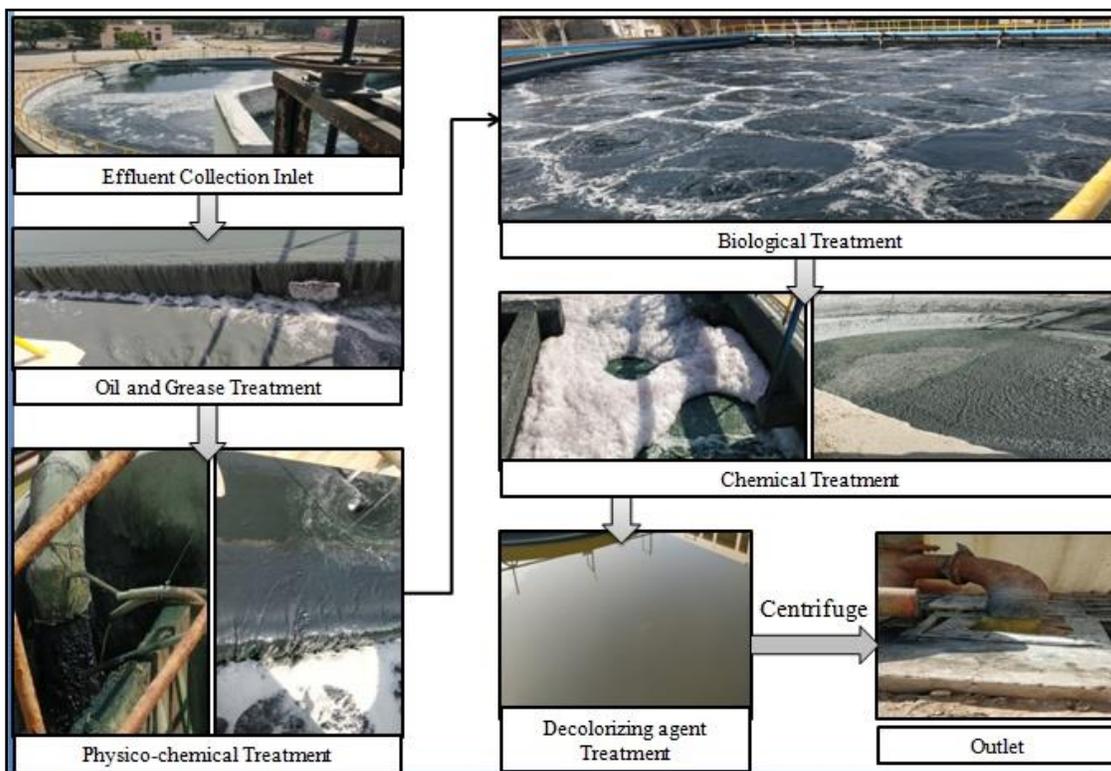


Fig. 1. Flowchart depicting the effluent flow inside an ETP through different treatment and its release through outlet after treatment

Bioremediation Approach (Microbial)

Biological treatments fundamentally rely on the ability of microbe to transform the contaminants by using them as sources of energy, carbon, and other minerals that are essential for their growth. Microbial-based enzymatic treatment is preferred for the degradation of the xenobiotic and recalcitrant azo dyes from the textile effluent because of the following advantages: (1) environmentally benevolent, (2) economic factors, (3) produces less sludge, (4) yielding end products that are non-toxic or have complete mineralization, and (5) requiring less water consumption as compared to the physicochemical methods. Thus, the challenge is to find microorganisms endowed with potential to degrade all azo dyes and at the same time thrive in the presence of salts, metals, other toxicants, and atypical conditions of textile effluents (Jamee and Siddique 2019).

Microbial nanoparticle approach

All the methods applied for wastewater treatment have different advantages and disadvantages. Nanoparticles, however, have gained attention due to their small size range (1 nm to 100 nm), large surface area, high adsorption properties, less resistance to diffusion and the fact that they show faster rates of equilibrium and increased chemical reactivity (Ahmad *et al.* 2015). Nanoparticle are divided into four functional classes that are used in water purification: carbonaceous nanomaterials, dendrimers, metal-containing nanoparticles and zeolites (Marimuthu *et al.* 2020). Nanotechnology-derived products reduce the level of toxic substances to sub-ppb levels and help attain higher water quality standards (Savage and Diallo 2005). Nanoparticle degrades or decolorizes azo dye either through absorption, photocatalytic degradation or their combined action. Nano-sized metal

oxides are preferred adsorbents for the removal of water toxins as such materials are associated with the characteristics of simplicity, efficacy, versatility, and high surface reactivity (Zafar *et al.* 2019). Photocatalysis is a principal mechanism in dye effluent treatment; here the electrons are excited from the valence band to conduction band upon irradiation, resulting in electron-hole pair generation. The hydroxyl radical generated acts as a potent oxidizing agent and completely degrades the dye to nontoxic products.

Nano-scale size provides properties such as improved catalysis, adsorption, and high reactivity. These properties have been exploited in recent years in all the domains, including wastewater treatment. Cruz *et al.* (2019) reported that cobalt nanoparticles (CoNPs) removed the Remazol golden yellow RNL by almost 100% in 30 min. The X-ray diffraction (XRD) and Raman spectroscopy study showed presence of CoO and Co⁰, which was supported by thermogravimetric analysis coupled to mass spectrometry (TG-MS) analysis. The application of CoNPs to textile effluent resulted in 88% degradation of dyes and 32% reduction in COD. Foster *et al.* (2019) used bimetallic nanoparticles comprising of iron (Fe) and nickel (Ni) at 1000 mg/L concentration, which showed high efficacy and consistent Orange G removal. Amabye and Hagos (2017) synthesized AgNPs using cell-free supernatant of an isolated bacterial strain AN-1 for the decolorization of dyes. Further characterization with scanning electron microscope (SEM) analysis revealed the spherical, polydisperse AgNPs of particle size ranging from 74.56 to 92.67 nm. Another study synthesized exopolysaccharide-stabilized AgNPs and characterized it using surface plasmon spectra using UV–VIS spectroscopy, XRD, TEM, SEM, atomic force microscopy (AFM), and Raman spectroscopy for MO and CR (Saravanan *et al.* 2017). Ramalingam *et al.* (2017) synthesized AgNPs using cell-free extract of *Staphylococcus aureus*. Roughly 62% of MO (2000 µg/mL) was degraded after treatment with 200 µg/mL of AgNPs. Nadaf and Kanase (2016) reported that gold nanoparticles (AuNPs) synthesized using cell-free extract of *Bacillus marisflavi* showed outstanding catalytic activity in the decolorization of CR and methylene blue. Modi *et al.* (2015) focused on synthesis of AgNPs using *Bacillus pumilus* and verified that nano-based remediation was more efficient as compared to microbial remediation (Table 7).

Table 7. Degradation of Azo Dyes Using Microbe Nanoparticle Approach

S.No.	Nanoparticles	Dye	Microbes	Remarks	References
1.	Cobalt nanoparticles (CoNPs)	Remazol golden yellow RNL; textile effluent	-	CoNPs removed the RGY with high efficiency, reaching almost 100% removal in 30min; textile effluent resulted in 88% degradation of dyes and 32% reduction in CO	Cruz <i>et al.</i> 2019
2.	Bimetallic nanoparticles comprising of iron (Fe) and nickel (Ni)	Orange G	-	High efficacy at 1000 mg/L	Foster <i>et al.</i> 2019
3.	Silver nanoparticles	Methylene blue	Strain AN-1	Promising agents for treatment of synthetic dyes	Amabye and Hagos 2017

4.	Silver nanoparticles	Congo Red, Rhodamine B, and Orange G	<i>Pestalotiopsis versicolor</i>	Good azo dye-degrading potential	Rajput <i>et al.</i> 2017
5.	Silver nanoparticles	Methyl Orange and Congo Red	<i>Leuconostoc lactis</i>	Exopolysaccharide-stabilized (EPS) AgNPs efficient in degradation process of industrial textile dyes; the electron transfer takes place from reducing agent to dye molecule <i>via</i> nanoparticles, resulting in the destruction of the dye chromophore structure	Saravanan <i>et al.</i> 2017
6.	Silver nanoparticles	Methyl Orange	<i>Staphylococcus aureus</i>	62% of MO (2000µg/mL) degraded with 200µg/mL of AgNPs in 1 day	Ramalingam <i>et al.</i> 2017
7.	Gold nanoparticles	Congo Red and Methylene blue	<i>Bacillus marisflavi</i>	Outstanding catalytic activity; pseudo-first-order kinetics and reaction rate constant of 0.2192 and 0.2484 min ⁻¹ , respectively	Nadaf and Kanase 2016
8.	Silver nanoparticles	Congo Red	<i>Bacillus pumillus</i>	Nano-based bioremediation was found 13% more efficient than the microbial remediation	Modi <i>et al.</i> 2015

CONCLUDING REMARKS

Discharge of untreated textile effluents in the natural environment is a widespread problem, especially where these industries are prominent. Treatment of textile wastewater is challenging, as it contains various toxic compounds possessing low biodegradability. Considering the vast metabolic and genetic diversity of microbial world and their role in dealing with various toxic compounds present in textile wastewater, there is much that remains unexplored. The authors wish to tap the unexplored microflora-based approach to find the best possible solution. Textile industries in India release a huge volume of effluent containing untreated or treated textile dye that is discharged into various drains adjoining the textile printing units. Elucidating structure, function, and diversity of the microbial community in these contaminated sites would facilitate the understanding of the biological process itself (tolerant as well as degrading). Investigation on structural as well as

functional diversity of indigenous microbes in contaminated sites is required to be explored, as such traits facilitate microbial metabolism during subsequent bioremediation activities in these environments. In particular, for industrial waste sites, the success of *in situ* remediation efforts could be critically monitored by studying the physiology and abundance of desirable bacteria/fungi by targeting the functional genes of indigenous microbial population.

In contrast, nanoparticles represent a promising new technology for environmental clean-up technology, not only because of their high treatment efficiency, but also for their cost-effectiveness, as they have the flexibility for *in situ* and *ex situ* applications. Nanoparticles work by increasing the surface area of a heterogeneous catalyst (its active component), enhancing the contact between the catalytic site and its substrate moiety. Efficacy of microbe-associated nanoparticle in bioremediation and its translational effects are unexploited. Thus, recovery of environmentally relevant microorganisms in combination with the 'omics' concept would facilitate an improved understanding of the physiology of microorganisms together with their nanoparticle synthesis catalyzing environmental processes. This increased understanding will further help to design and operate relevant bioremediation strategies.

ACKNOWLEDGEMENTS

The authors are grateful to Professor Aditya Shastri, Vice-Chancellor Banasthali Vidyapith, Rajasthan. The authors also give thanks to DST-CURIE for providing financial assistance in conducting our research work. The author wish to acknowledge the support from the Department of Science and Technology for providing INSPIRE fellowship for the project.

REFERENCES CITED

- Adebajo, S. O., Balogun, S. A., and Akintokun, A. K. (2017). "Decolourization of vat dyes by bacterial isolates recovered from local textile mills in Southwest, Nigeria," *Microbiol. Res. J. Int.* 18(1), 1-8. DOI: 10.9734/MRJI/2017/29656
- Ahmad, A., Mohd-Setapar, S. H., Chuong, C. S., Khatoon, A., Wani, W. A., Kumar, R., and Rafatullah, M. (2015). "Recent advances in new generation dye removal technologies: novel search for approaches to reprocess wastewater," *RSC Adv.* 5(39), 30801-30818. DOI: 10.1039/C4RA16959J
- Ahmed, F., Dewani, R., Pervez, M. K., Mahboob, S. J., and Soomro, S. A. (2016). "Non-destructive FT-IR analysis of mono azo dyes," *Bulg. Chem. Commun.* 48(1), 71-77.
- Ajaz, M., Rehman, A., Khan, Z., Nisar, M. A., and Hussain, S. (2019). "Degradation of azo dyes by *Alcaligenes aquatilis* 3c and its potential use in the wastewater treatment," *AMBExpr.* 9(64), 1-12. DOI: 10.1186/s13568-019-0788-3
- Allam, N. G. (2017). "Bioremediation efficiency of heavy metals and azo dyes by individual or consortium bacterial species either as free or immobilized cells: A comparative study," *Egypt. J. Bot.* 57(3), 555-564. DOI: 10.21608/ejbo.2017.689.1040
- Almeida, E. J. R., and 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. DOI: 10.1016/j.chemosphere.2014.04.060
- Al-Tohamy, R., Kenawy, E. R., Sun, J., and Ali, S. S. (2020). “Performance of a newly isolated salt-tolerant yeast strain *Sterigmatomyces halophilus* SSA-1575 for azo dye decolorization and detoxification,” *Front. Microbiol.* 11(1163). DOI: 10.3389/fmicb.2020.01163
- Amabye, T. G., and Hagos, K. (2017). “Green synthesis of silver nanoparticles and evaluation of its potential for decolourization of a synthetic dye and antibacterial activity,” *Int. J. Nano. Med. Eng.* 2(7), 120-126. DOI: 10.25141/2474-8811-2017-7.0120
- American Public Health Association (APHA) (1998). *Standard Methods for the Examination of Water and Wastewater*, 20th Edition, Water and Environment Federation and American Water Works Association, Washington, D.C., USA.
- Asses, N., Ayed, L., Hkiri, N., and Hamdi, M. (2018). “Congo Red decolorization and detoxification by *Aspergillus niger*: Removal mechanisms and dye degradation pathway,” *Biomed Res. Int.* 2018, Article ID 3049686. DOI: 10.1155/2018/3049686
- Ayed, L., Mahdhi, A., Cheref, A., and Bakhrouf, A. (2011). “Decolorization and degradation of azo-dye Methyl Red by an isolated *Sphingomonas paucimobilis*: Biototoxicity and metabolites characterization,” *Desalination* 274(1-3), 272-277. DOI: 10.1016/j.desal.2011.02.024
- Baban, A., Yediler, A., Lienert, D., Kemerdere, N., and Kettrup, A. (2003). “Ozonation of high strength segregated effluents from a woollen textile dyeing and finishing plant,” *Dyes Pigm.* 58(2), 93-98. DOI: 10.1016/S0143-7208(03)00047-0
- Banat, I. M., Nigam, P., Singh, D., and Marchant, R. (1996). “Microbial decolorization of textile-dye- containing effluents: A review,” *Bioresource Technol.* 58(3), 217-227. DOI:10.1016/S0960-8524(96)00113-7
- Carmen, Z., and Daniela, S. (2012). “Textile organic dyes – Characteristics, polluting effects and separation/elimination procedures from industrial effluents – A critical overview,” in: *Organic Pollutants Ten Years After the Stockholm Convention- Environmental and Analytical Update*, Tomasz Puzyn and Aleksandra Mostrag-Szlichtyng (eds.), InTechOpen, pp. 55-86. DOI: 10.5772/32373
- Cheng, W. N., Sim, H. K., Ahmad, S. A., Syed, M. A., Shukor, M. Y., and Yusof, M. T. (2016). “Characterization of an azo-dye-degrading white rot fungus isolated from Malaysia,” *Mycosphere* 7(5), 560-569. DOI: 10.5943/mycosphere/7/5/3
- Cruz, J. C., Nascimento, M. A., Amaral, H. A. V., Lima, D. S. D., Teixeira, A. P. C., and Lopes, R. P. (2019). “Synthesis and characterization of cobalt nanoparticles for application in the removal of textile dye,” *J. Environ. Manage.* 242, 220-228. DOI: 10.1016/j.jenvman.2019.04.059
- Dafale, N., Agrawal, L., Kapley, A., Meshram, S., Purohit, H., and Wate, S. (2010). “Selection of indicator bacteria based on screening of 16s rDNA metagenomic library from a two-stage anoxic-oxic bioreactor system degrading azo dyes,” *Bioresource Technol.* 101(2), 476-484. DOI: 10.1016/j.biortech.2009.08.006
- Desai, S. A. (2017). “Isolation and characterization dye degrading bacteria for detoxification of dark red 2B,” *Biosci. Discov.* 8(3), 426-431.
- Elango, G., Rathika, G., and Elango, S. (2017). “Physico-chemical parameters of textile dyeing effluent and its impacts with case study,” *Int. J. Res. Chem. Environ.* 7(1), 17-24.

- El-Sheekh, M. M., Gharieb, M. M., and Abou-El-Souod, G. W. (2009). "Biodegradation of dyes by some green algae and cyanobacteria," *Int. Biodeterior. Biodegradation* 63 (2009), 699-704. DOI: 10.1016/j.ibiod.2009.04.010
- Fatima, N., and Alamgir, A. (2015). "Decolourization of textile effluent through indigenous microorganism," *Bull. Env. Pharmacol. Life Sci.* 4(5), 109-115. DOI: 10.13140/RG.2.2.17154.53442
- Foster, S. L., Estoque, K., Voecks, M., Rentz, N., and Greenlee, L. F. (2019). "Removal of synthetic azo dye using bimetallic nickel-iron nanoparticles," *J. Nanomater.* 2019, Article ID 9807605. DOI: 10.1155/2019/9807605
- Gita, S., Shukla, S. P., Prakash, C., Saharan, N., and Deshmukhe, G. (2018). "Evaluation of toxicity of a textile dye (Optilan Red) towards a green microalga *Chlorella vulgaris*," *Int. J. Curr. Microbiol. App. Sci.* 7(8), 3346-3355. DOI: 10.20546/ijcmas.2018.708.358
- Hasan, M. K., and Miah, M. (2014). "Impacts of textile dyeing industries effluents on surface water quality: A study on Araihaazar Thana in Narayanganj District of Bangladesh," *J. Environ. Human.* 1(3), 8-22. DOI:10.15764/EH.2014.03002
- Hernandez-Zamora, M., Cristiani-Urbani, E., Martinez-Jeromino, F., Perales-Vela, H. V., Ponce-Noyola, T., Montes-Horcasitas, M. C., and Canizares-Villanueva, R. O. (2015). "Bioremoval of the azo dye Congo Red by the microalga *Chlorella vulgaris*," *Environ. Sci. Pollut. Res.* 22(14), 10811-10823. DOI 10.1007/s11356-015-4277-1
- Hossen, M. Z., Hussain, M. E., Hakim, A., Islam, K., Uddin, M. N., and Azad, A. K. (2019). "Biodegradation of reactive textile dye Novacron Super Black G by free cells of newly isolated *Alcaligenes faecalis* AZ26 and *Bacillus* spp obtained from textile effluent," *Heliyon* 5(7), 1-11. DOI: 10.1016/j.heliyon.2019.e02068
- Ishchi, T., and Sibi, G. (2020). "Azo dye degradation by *Chlorella vulgaris*: Optimization and kinetics," *Int. J. Biol. Chem.* 14 (1), 1-7. DOI: 10.3923/ijbc.2020.1.7
- Islam, T., Rahman, M. S., and Hussain, M. S. (2017). "Heavy metal tolerance pattern of textile dye degrading native bacteria: A bioremediation viewpoint," *Ann. Med. Health Sci. Res.* 7(1), 67-73.
- Jaishree, and Khan, T. I. (2014). "Monitoring of heavy metals in textile waste water of Sanganer, Jaipur (Rajasthan)," *IJSPR* 4(3), 1-4.
- Jamee, R., and Siddique, R. (2019). "Biodegradation of synthetic dyes of textile effluent by microorganisms: An environmentally and economically sustainable approach," *Eur. J. Microbiol. Immunol.* 9(4), 114-118. DOI: 10.1556/1886.2019.00018
- Kalyani, D. C., Patil, P. S., Jadhav, J. P., and Govindwar, S. P. (2008). "Biodegradation of reactive textile dye red BLI by an isolated bacterium *Pseudomonas* sp. SUK1," *Bioresource Technol.* 99(11), 4635-4641. DOI: 10.1016/j.biortech.2007.06.058
- Karunya, A., Rose, C., and Valli Nachiyar, C. (2014). "Biodegradation of the textile dye Mordant Black 17 (Calcon) by *Moraxella osloensis* isolated from textile effluent-contaminated site," *World J. Microbiol. Biot.* 30(3), 915-924. DOI: 10.1007/s11274-013-1509-8
- Khan, S., and Malik, A. (2018). "Toxicity evaluation of textile effluents and role of native soil bacterium in biodegradation of a textile dye," *Environ. Sci. Pollut. Res.* 25(5), 4446-4458. DOI: 10.1007/s11356-017-0783-7
- Khandare, R. V., Kabra, A. N., Awate, A. V., and Govindwar, S. P. (2013). "Synergistic degradation of diazo dye Direct Red 5B by *Portulaca grandiflora* and *Pseudomonas putida*," *Int. J. Environ. Sci. Technol.* 10(5), 1039-1050. DOI: 10.1007/s13762-013-0244-x

- Khataee, A. R., Zarei, M., Dehghan, G., Ebadi, E., and Pourhassan, M. (2010). "Biotreatment of a triphenylmethane dye solution using a *Xanthophyta* alga: Modeling of key factors by neural network," *J. Taiwan Inst. Chem. Eng.* 42(2011), 380-386. DOI: 10.1016/j.jtice.2010.08.006
- Khatun, S. (2017). "Toxicity impact of silk dye waste effluent induced histopathological changes in the stomach and intestine of Swiss albino mice *Mus musculus* and their mitigation using *Moringa oleifera* leaf powder," *J. Med. Sci. Clin. Res.* 5(3), 19306-19313. DOI: 10.18535/jmscr/v5i3.156
- Lade, H., Govindwar, S. P., and Paul, D. (2015). "Low-cost biodegradation and detoxification of textile azo dye C.I. Reactive Blue 172 by *Providencia rettgeri* strain HSL1," *J. Chem.* 2015, Article ID 894109. DOI: 10.1155/2015/894109
- Lalnunhlumi, S., and Krishnaswamy, V. (2016). "Decolorization of azo dyes (Direct Blue 151 and Direct Red 31) by moderately alkaliphilic bacterial consortium," *Braz. J. Microbiol.* 47(1), 39-46. DOI: 10.1016/j.bjm.2015.11.013
- Laxmi, S., and Nikam, T.D. (2015). "Decolorisation and detoxification of widely used azo dyes by fungal species isolated from textile dye contaminated site," *Int. J. Curr. Microbiol. App. Sci.* 4(4), 813-834.
- Lobiuc, A., Olaru, S., Hancu, E. I., Costica, N., Fortuna, M. E., Zamfirache, M. M., and Constantinescu, G. (2018). "Toxicity and removal of Direct Red 28 diazo dye in living polymeric systems," *Rev. Chim. (Bucharest)* 69(7), 1628-1635. DOI: 10.37358/RC.18.7.6384
- Mahawar, P., and Akhtar, A. (2015). "Physico-chemical characterization of soil and effluent of dye industries in Kaithun region of Kota, Rajasthan," *Int. J. Pure App. Biosci.* 3(2), 419-422.
- Marimuthu, S., Antonisamy, A. J., Malayandi, S., Rajendran, K., Tsai, P. C., Pugazhendhi, A., and Ponnusamy, V. K. (2020). "Silver nanoparticles in dye effluent treatment: A review on synthesis, treatment methods, mechanisms, photocatalytic degradation, toxic effects and mitigation of toxicity," *J. Photochem. Photobiol. B.* 205(111823). DOI: 10.1016/j.jphotobiol.2020.111823
- Modi, S., Pathak, B., and Fulekar, M. H. (2015). "Microbial synthesized silver nanoparticles for decolorization and biodegradation of azo dye compound," *J. Environ. Nanotechnol.* 4(2), 37-46. DOI: 10.13074/jent.2015.06.152149
- Nadaf, N., and Kanase, S. S. (2016). "Biosynthesis of gold nanoparticles by *Bacillus marisflavi* and its potential in catalytic dye degradation," *Arab. J. Chem.* 12(8), 4806-4814. DOI: 10.1016/j.arabjc.2016.09.020
- Ozkan, Z. Y., Cakirgoz, M., Kaymak, E. S., and Erdim, E. (2018). "Rapid decolorization of textile wastewater by green synthesized iron nanoparticles," *Water Sci. Technol.* 77(2), 511-517. DOI: 10.2166/wst.2017.559
- Parmar, N. D., and Shukla, S. R. (2018). "Decolourization of dye wastewater by microbial methods – A review," *Indian J. Chem. Technol.* 25(4), 315-323.
- Patel, R., Tajddin, K., Patel, A., and Patel, B. (2015). "Physico-chemical analysis of textile effluent," *IJRSI* 2(5), 33-37.
- Przystaś, W., Zabłocka-Godlewska, E., and Grabińska-Sota, E. (2012). "Biological removal of azo and triphenylmethane dyes and toxicity of process by-products," *Water Air Soil Poll.* 223(4), 1581-1592. DOI: 10.1007/s11270-011-0966-7
- Rahi, R. K., Prasad, R. N., and Gupta, V. (2018). "Analysis of physico-chemical properties of textile effluents collected from Sanganer, Jaipur," *Int. J. Adv. Sci. Res. Manag.* 3(7), 119-123.

- Rajeswari, K., Subashkumar, R., and Vijayaraman, K. (2014). "Degradation of textile dyes by isolated *Lysinibacillus Sphaericus* strain RSV-1 and *Stenotrophomonas maltophilia* strain RSV-2 and toxicity assessment of degraded product," *J. Environ. Anal. Toxicol.* 4(4), 222-226. DOI: 10.4172/2161-0525.1000222
- Rajput, K., Agrawal, S., Sharma, J., and Agrawal, P. K. (2017). "Mycosynthesis of silver nanoparticles using *Pestalotiopsis versicolor* endophytic fungus and investigation of its antibacterial and azo dye degradation efficacy," *Kavaka* 49, 65-71.
- Ramalingam, V., Dhanasundari, S., Nithiya, P., and Rajaram, R. (2017). "Catalytic degradation of Methyl Orange using biogenic nanosilver and its phytotoxicity evaluation," *Indian J. Chem. Technol.* 24(3), 336-343.
- Rani, B., Kumar, V., Singh, J., Bisht, S., Teotia, P., Sharma, S., and Kela, R. (2014). "Bioremediation of dyes by fungi isolated from contaminated dye effluent sites for bio-usability," *Braz. J. Microbiol.* 45(3), 1055-1063. DOI: 10.1590/s1517-83822014000300039
- Sani, Z. M., Abdullahi, I. L., and Sani, A. (2018). "Toxicity evaluation of selected dyes commonly used for clothing materials in urban Kano, Nigeria," *Eur. J. Exp. Biol.* 8(4), 1-4. DOI: 10.21767/2248-9215.100067
- Saravanan, C., Rajesh, R., Kaviarasan, C., Muthukumar, K., Kavitate, D., and Shetty, P. H. (2017). "Synthesis of silver nanoparticles using bacterial exopolysaccharide and its application for degradation of azo-dyes," *Biotechnol. Rep.* 15, 33-40. DOI: 10.1016/j.btre.2017.02.006
- Sarkar, S., Banerjee, A., Halder, U., Biswas, R., and Bandopadhyay, R. (2017). "Degradation of synthetic azo dyes of textile industry: A sustainable approach using microbialenzymes," *Water Conserv. Sci. Eng.* 2, 121-131. DOI: 10.1007/s41101-017-0031-5
- Satija, A., and Bhatnagar, M. (2017). "Environmental assessment of textile wastewater of Sanganer area in Jaipur," *Int. J. Innov. Res. Sci. Eng. Technol.* 6(6), 10779-10786. DOI:10.15680/IJIRSET.2017.0606116
- Savage, N., and Diallo, M. S. (2005). "Nanomaterials and water purification: Opportunities and challenges," *J. Nanopart. Res.* 7(4), 331-342. DOI: 10.1007/s11051-005-7523-5
- Sharma, S., Arti, K., Viswanadhappalli, S., Singh, P. K., Shipra, R., and Sharma, K. P. (2007). "Toxicity assessment of textile dye wastewater using Swiss albino rats," *Australas. J. Ecotoxicol.* 13(2), 81-85.
- Sharma, S., Verma, C. S., Singh, A. K., Bajpai, K., and Singh, S. K. (2013). "Use of metagenomic approach to diversify bacteria from textile effluent contaminated soil," *Int. J. Sci. Eng. Res.* 4(11), 1773-1781.
- Singh, A. L., Chaudhary, S., and Yadav, A. (2017). "Decolourization, degradation and removal of heavy metals of textile effluent with the help of mixed bacterial consortium," *Indian J. Biotechnol.* 16(2), 258-264.
- Singh, R. P., Singh, P. K., and Singh, R. L. (2014). "Bacterial decolorization of textile azo dye Acid Orange by *Staphylococcus hominis* RMLRT03," *Toxicol. Int.* 21(2), 160-166. DOI:10.4103/0971-6580.139797
- Sponza, D. T., and Isak, M. (2004). "Toxicity and intermediates of C.I. Direct Red 28 dye through sequential anaerobic/aerobic treatment," *Process Biochem.* 40(8), 2735-2744. DOI:10.1016/j.procbio.2004.12.016
- Sriram, N., and Reetha, D. (2015). "Isolation and characterization of dye degrading bacteria from textile dye effluents," *Cent. Eur. J. Exp. Biol.* 4(2), 5-10.

- Starovoitova, D., and Odido, D. (2014). "Assessment of toxicity of textile dyes and chemicals *via* materials safety data sheets," *RRBS* 9(7), 241-248.
- Sudha, M., Saranya, A., Selvakumar, G., and Sivakumar, N. (2014). "Microbial degradation of azo dyes: A review," *Int. J. Curr. Microbiol. Appl. Sci.* 3(2), 670-690.
- Tang, W., Xu, X., Ye, B.C., Cao, P., and Ali, A. (2019). "Decolorization and degradation analysis of Disperse Red 3B by a consortium of the fungus *Aspergillus* sp. XJ-2 and the microalgae *Chlorella sorokiniana* XJK," *RSC Adv.* 9(25), 14558-14566. DOI: 10.1039/c9ra01169b
- Vimala, G., Jeyakumar, P., Devi. C. A., Singh, A., and Iyer, P. (2015). "Azo dye degrading bacteria from textile effluent," *Int. J. Curr. Microbiol. Appl. Sci.* 4(7), 199-210.
- Yang, P., Shi, W., Wang, H., and Liu, H. (2016). "Screening of freshwater fungi for decolorizing multiple synthetic dyes," *Braz. J. Microbiol.* 47(4), 828-834. DOI: 10.1016/j.bjm.2016.06.010
- Zafar, M. N., Dar, Q., Nawaz, F., Zafar, M. N., Iqbal, M., and Nazar, M. F. (2019). "Effective adsorptive removal of azo dyes overspherical ZnO nanoparticles," *J. Mater. Res. Technol.* 8(1), 713-725. DOI: 10.1016/j.jmrt.2018.06.002
- Zhang, W., Liu, W., Zhang, J., Zhao, H., Zhang, Y., Quan, X., and Jin, Y. (2012). "Characterisation of acute toxicity, genotoxicity and oxidative stress posed by textile effluent on zebrafish," *J. Environ. Sci.* 24(11), 2019-2027. DOI: 10.1016/S1001-0742(11)61030-9

Article submitted: May 21, 2020; Peer review completed: August 23, 2020; Revised version received and accepted: September 14, 2020; Published: September 21, 2020. DOI: 10.15376/biores.15.4.Saxena