

Review

A Review of Various Treatment Methods for the Removal of Dyes from Textile Effluent

Chingrishon Kathing[†], Geeta Saini^{†, *}Department of Chemistry, Gargi College, University of Delhi, India; E-Mails:
Chingrishon.kathing@gargi.du.ac.in; Geeta.saini@gargi.du.ac.in[†] These authors contributed equally to this work.^{*} **Correspondence:** Geeta Saini; E-Mail: Geeta.saini@gargi.du.ac.in**Academic Editors:** Laila Mandi and Miquel Salgot**Special Issue:** [Wastewater Reclamation and Reuse](#)*Recent Progress in Materials*
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Abstract

Wastewater generated by the textile industry has been a major environmental concern for a long. Production of fiber involves various steps and uses a lot of chemicals, dyes, and water. Therefore, the effluent produced from the textile industry needs proper purification before discharging into the water body. The current review summarizes various physical and chemical methods like ion exchange, coagulation-flocculation, membrane separation, membrane distillation, oxidation, ozonation, etc., for wastewater treatment. Along with this, adsorption methods, the various adsorbents used to purify wastewater, and the mechanism involved in adsorption have also been discussed. The biological method utilizes various microbes (bacteria, fungi, algae, and yeast) as a whole and the enzymes (laccase and azoreductase) secreted by them for wastewater treatment, which have been considered more feasible than physical and chemical methods. The adsorption and biological methods are better than other techniques due to their ability to degrade diverse classes of dye, less accumulation of harmless sludge, and cost-effective and safer approach for the disposal of textile effluent. While physical and chemical methods are expensive and generate toxic sludge, which is difficult to decompose.



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Keywords

Textile industry; wastewater; treatment methods; biosorption; physical method; chemical method

1. Introduction

Water is essential to life. It is required for survival, making it one of the most important and potent elements of life for all living things. Unfortunately, rapid industrialization, modernization, and various anthropogenic activities have contaminated water bodies. The textile industry has also become one of the biggest environmental challenges, despite playing a key part in meeting people's basic needs since the beginning [1-3]. It has been estimated that textile dyeing contributed nearly 20% of global wastewater, or around 93 billion cubic meters producing highly colored wastewater [4]. About 7×10^7 tons of synthetic dyes are produced annually worldwide, with over 10,000 tons of such dyes by textile industries [5]. In addition to being one of the most polluting industries, it is also the largest consumer of water, resulting in water depletion and a threat to water scarcity. Directly discharging untreated wastewater containing dyes into water bodies creates pollution that ultimately damages water quality and causes major health problems in humans [6, 7]. Several reports are available in the literature to prove the health hazard related to dyes [8, 9]. The minute amount of metals discharged by textile effluents is toxic to animals and humans [10, 11]. The highly contaminated water enters into the groundwater, which deteriorates the water quality of groundwater. Textile dyes are highly carcinogenic and may cause various skin and eye ailments in humans. e.g., dermatitis, allergic conjunctivitis, and occupational asthma are common diseases caused by textile dyes [12, 13]. The most probable reason for the ailments caused by textile dyes is the replacement or denaturation of enzymatic activity [14]. In addition, it degrades the aquatic ecosystem, as the dye in the water reduces its transparency, inhibits photosynthesis, and subsequently lowers dissolved oxygen (D.O.), harming both aquatic flora and fauna [15]. At decreased D.O. level, the microorganism could not survive, and the natural process of decomposition of organic waste by bacteria and other microorganisms does not occur, which results in the accumulation of organic waste in the water body. Pollutants such as dyes, various organic and inorganic compounds, and heavy metals such as Hg, Cr, Pb, and As, which are poisonous by nature and represent significant environmental issues, are prevalent in textile industry wastewater [16-18].

The production of fabric from textile fiber involves many steps, as shown in Figure 1, and substantial amounts of chemicals are used at each step, further increasing water pollution [19]. Dye is the primary pollutant in textile effluent water [20]. Dye effluents are generated because dyes are not completely fixed to the fiber during the dyeing and finishing operations [21, 22]. Dye molecules have a complicated aromatic backbone and contain several auxochromes that give them a beautiful color. Therefore, they have a very complex and stable structure that poses greater difficulty in breakdown and degradation; hence removal from textile effluent becomes more challenging [23].

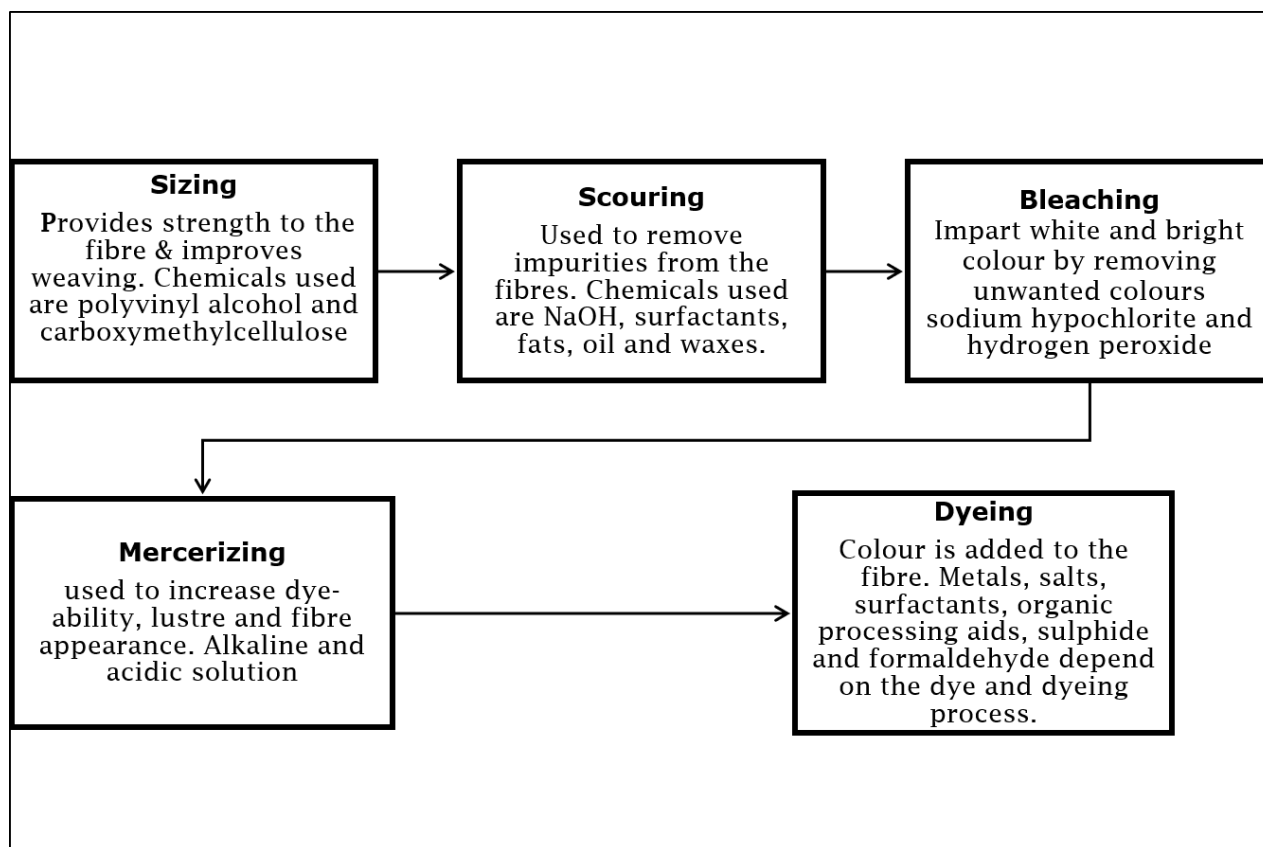


Figure 1 Various processes occur in the production of fabric from the fiber.

There are various categories of dyes based on origin, structure, and application, such as acidic, basic, disperse, direct, reactive, azo, diazo, and mordant dye, summarized in Figure 2 [24].

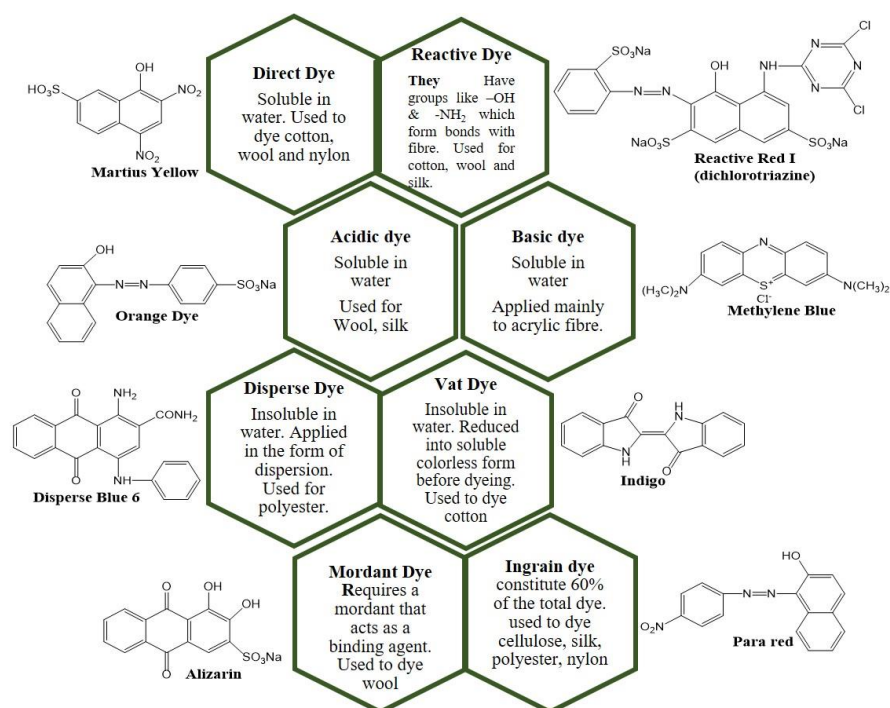


Figure 2 Classification of dyes.

Azo dye represents the greatest production volume in dyestuff chemicals today, and their relative importance may grow. These are extensively used in the textile, paper, food, leather, cosmetics, and pharmaceutical industries and account for about 50 % of all dyes synthesized [25].

Due to the ease of coupling reactions, it is possible to synthesize the desired dye, which is suitable for a wide range of applications [26]. The intricate structure of dyes prevents their degradation; thus, they remain in the water for longer. For instance, the half-life of hydrolyzed Reactive Blue 19 is about 46 years at pH 7 and 25°C [27]. Since most of the azo dyes under anaerobic conditions decomposed as aromatic amines, which can act as carcinogens and mutagens, their removal from wastewater before discharge into water bodies is of utmost importance and is a challenging task to formulate high-efficiency treatment methods [28, 29]. The treatment of wastewater to make them suitable for subsequent use requires physical, chemical, and biological processes, which are depicted in Figure 3 [30]. In the textile industry, selecting the most efficient and cost-effective treatment processes or their combinations is contingent on the dyestuffs and dyeing techniques employed in production [27, 31-33].

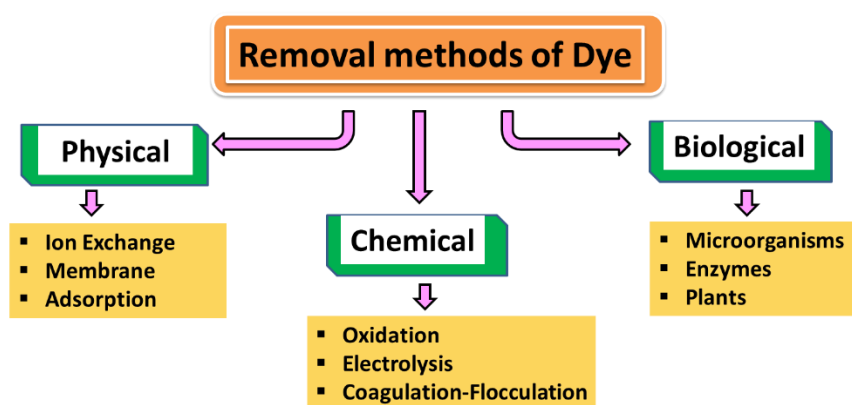


Figure 3 Various treatment methods used for the purification of textile effluents.

1.1 Physical Methods

Various physical methods, such as ion exchange [34], membrane separation [35, 36], and adsorption [32], etc., have been employed for many years for the treatment of wastewater [37]. Ion exchange is a well-developed technique involving an exchange of mobile ions from an external solution that is electrostatically bound to the functional groups within a solid matrix [38]. It is used for purifying, separating, and decontamination of aqueous and other ion-containing solutions [39]. Despite its wide applications, there are limitations associated with the ion exchange method as it generates a highly concentrated waste that requires careful disposal. Generally, it is ineffective to treat all categories of dyes and for wastewater containing high concentrations of metal ions such as Fe, Mn, Al, etc. Membrane separation processes such as reverse osmosis (RO) [40], nanofiltration (NF) [41], and microfiltration (MF) [42] have also been successfully employed in the treatment of dye-laden water (more than or about 90% removal efficiency). However, pore blockage and membrane fouling during these processes remain significant limitations [43].

In recent times, membrane distillation (MD) technology has emerged as the most desirable and effective method for wastewater reclamation. The most important feature of membrane distillation is the ability to pass water vapor (volatile parts) through the microporous hydrophobic membrane

pores and collect as distillate in a pure form [44]. Some of the benefits offered by MD are the significant reduction of equipment size, low operating temperature and pressure, low energy requirement, and low capital cost. Despite its excellent performance and sought-after method, pore wetting, membrane fouling, and scaling are the drawbacks encountered, particularly with the membrane distillation method [45].

MD processes can be classified into four major configurations depending on the methods to induce a vapor pressure difference across the membrane and the type of permeate collection on the cold side, summarized in Table 1.

Table 1 Types of Membrane Distillation (MD) Configuration and advantages and disadvantages.

S.no	Types of Membrane (MD) Configuration	Advantage	Disadvantage
1	Direct Contact MD (DCMD): Both feed and permeate solutions are in direct contact with the membrane [46]	<ul style="list-style-type: none"> • High rejection of non-volatile • Moderate operating temperatures • Less sensitive to feed concentration • Low maintenance and operation cost [47] 	<ul style="list-style-type: none"> • The cold feed cannot be used as a coolant • High heat loss by conduction [48]
2	Air Gap MD (AGMD): The hot feed solution is in direct contact with the hydrophobic surface of the membrane, while an additional air gap separates the permeate side of the membrane and a condensation surface [46]	<ul style="list-style-type: none"> • Low conductive heat loss • Low membrane wetting [48] 	<ul style="list-style-type: none"> • Low permeate flux [48]
3	Sweeping Gas MD (SGMD): A cold sweep gas provides the driving force in sweep gas membrane distillation [46] [49]	<ul style="list-style-type: none"> • Reduce conductive heat loss [48] 	<ul style="list-style-type: none"> • Requires a big condenser because of the large volume of the sweep gas [48]
4	Vacuum MD (VMD): Vacuum pressure is applied at the permeate side of the membrane to collect the vapors, and vapor condensation takes place outside the membrane module [50, 51]	<ul style="list-style-type: none"> • High permeate flux • Cost-effective • Low thermal and conductive heat loss • Remove volatile organic compounds [50, 52] 	<ul style="list-style-type: none"> • Susceptible to wetting severely due to the applied vacuum pressure [48]

1.2 Chemical Methods

Chemical method of wastewater treatment includes oxidation, electrolysis, and coagulation-flocculation methods [53]. The use of advanced oxidation processes (AOP) [54] such as ozonation [55], ultraviolet/hydrogen peroxide (UV/H₂O₂) [56, 57], Fenton, [58, 59] ultrasound [60], anodic oxidation [61], and photocatalytic processes [62] have been efficiently employed to treat dye wastewater.

The coagulation and flocculation method are physicochemical treatments used frequently for wastewater treatment with minimal generation of harmful and toxic intermediates. High molecular weights of surfactants and dyes are successfully removed by the coagulation and flocculation processes followed by sedimentation, flotation, and filtration, respectively [63]. Decolorization by this approach is achieved by eliminating dye molecules from dyebath effluents rather than through the partial decomposition of dyes, which could generate potentially hazardous and poisonous aromatic chemicals [64]. However, the method has drawbacks, such as the generation of inherent sludge and disposal problems [65]. Table 2 summarizes the different physical and chemical methods used to remove specific dyes.

Table 2 Physical and chemical methods used for removal of dyes.

Physical method		Chemical method	
Ion exchange	Methylene Blue	Oxidation	Acid Black 1 [66]
	Eosin Y		Direct Red 80 [67]
	Malachite Green		Acid Red 14 [68]
	Methyl Red [38]		Methyl Orange [69]
			Alizarin Red S [70]
			Crystal Violet [71]
Membrane distillation/separation	Acid Red, Reactive black, and	Electrolysis	Reactive Black B [72]
	Reactive Blue [40]		Methylene Blue [73]
	Direct Red 16,		C.I. Reactive Red 2 [74]
	Methylene blue [42]		Methyl Orange [75]
	Methyl orange [51]	Coagulation-flocculation	Acid Orange 7 [76]
			Eriochrome Black [77]
			Acid Red 119 [78]
			Direct Black 19, Direct Red 28, Direct Blue 86 [79]
			Congo Red [80]

1.3 Biological Methods

Biological treatment is considered the best method due to its ability to degrade diverse classes of dye, less accumulation of harmless sludge, and a cost-effective and safer approach to the disposal of textile effluent [81]. Bacterial degradation can occur *via* bacteria cells, enzymes present in

bacteria, and bacterial consortium, as displayed in Figure 4 [82]. A few examples of microbes used for degrading various types of dyes have been given in Table 1.

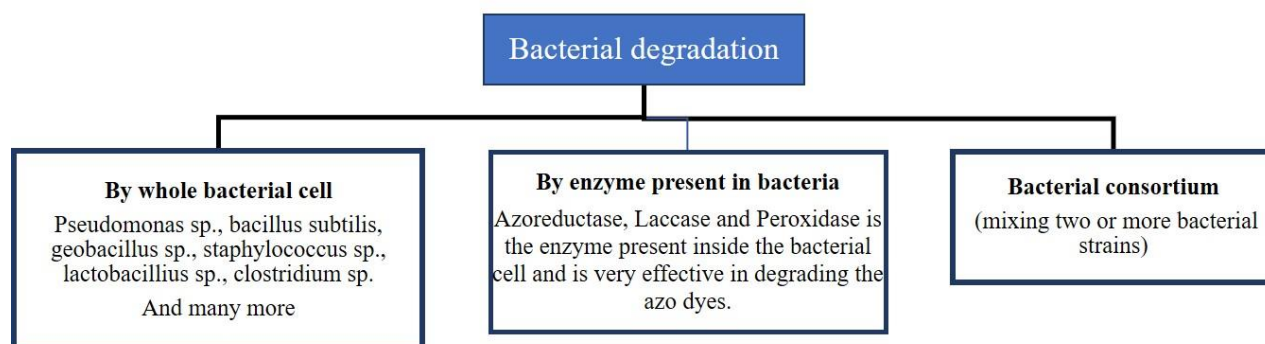


Figure 4 Various methods for bacterial degradation.

Azoreductase, laccase, and peroxidase are the enzymes produced by bacteria and have proven to be very effective in the degradation of complex dye molecules, particularly azo dye [83-85]. The microbial degradation of azo dyes to their corresponding amines is initiated by the cleavage of the azo linkage (-N=N-) with an anaerobic azo reductase [86]. Various Co-enzymes that reduce azo linkages are (Flavin Mononucleotide) FMN-independent reductases [87] and (Nicotinamide Adenine Dinucleotide Phosphate) NADPH-dependent reductases [88]. A colorless solution is obtained by reductive cleavage of the azo bond, which is converted into amines and other degraded products in the next step by oxidative cleavage, as depicted in Figure 5 [89]. Therefore, the enzymatic method has gained momentum in the last decade as it gives non-toxic end products, produces less sludge than other methods, and substantially increases the reaction rate, requiring much less time [90]. However, the amount of enzyme production is limited, and they are highly susceptible to undergoing deactivation by various physical and chemical parameters [91]. In addition, the method is overshadowed by limitations such as space requirements, low removal rate, and inefficiency in treating recalcitrant dye components [92, 93].

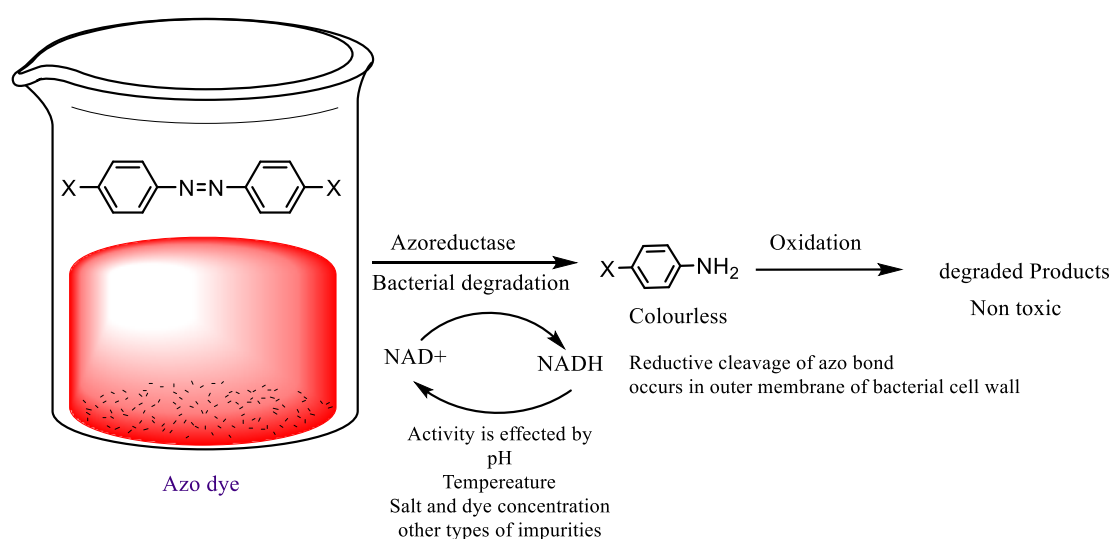


Figure 5 Schematic presentation of degradation of azo dye.

Mycoremediation, a biosorption by fungi, holds many benefits in the same way as the microbial process. The most widely used fungi in the decolorization and degradation of dye are the ligninolytic fungi of class basidiomycetes [94-96]. Algae with high surface area and binding affinity enable the removal of dye with high efficiency. Yeast-assisted biosorption is less explored, yet they are found to be superior and more sustainable mainly due to their ability to adapt to harsh environmental conditions [97, 98]. It has been studied that yeast species like *Saccharomyces cerevisiae*, *Saccharomyces uvarum*, *Saccharomyces lipolytic*, and *Turolopsis candida* can effectively degrade Reactive Brilliant Red K-2BP through biosorption [97]. Various microorganisms used for the degradation of dye molecule are indicated in Table 3.

The strong electrostatic force of attraction between the dye and functional groups like carboxyl, hydroxyl, phosphate, and other charged groups present in the microbes' cell walls governs the biosorption capacity assisted by microorganisms [99]. Furthermore, biosorption efficiency is regulated by parameters like pH, temperature, ionic strength, time of contact, adsorbent and dye concentration, dye structure, and type of microorganisms [100]. Moreover, the rapid growth rate of bacteria and simplicity in handling enables the process to be highly favored for removing dye from wastewater [101].

Table 3 Microbial degradation of a few selected dyes.

Microorganism	Species	Dye
Bacteria	<i>Corynebacterium glutamicum</i> [102-104]	Reactive Red 4, Reactive Yellow 2, Methylene Blue
	<i>Bacillus weihenstephanensis</i> RI12 [105]	Congo Red
	<i>Citrobacter</i> sp. CK3 [106]	Reactive Red 180
	<i>Rhizobium radiobacter</i> [107]	Reactive Red 141
	<i>Citrobacter</i> sp. <i>Pseudomonas</i> sp. [108]	Reactive Red 2
	<i>Enterococcus faecalis</i> Y, Z 66 [109]	Reactive Orange II
	<i>Bacillus lentus</i> BI 377 [110]	Reactive Red 120
	<i>Bacillus fusiformis</i> KMK5 [111]	Disperse Blue 79, Acid Orange 10
	<i>Kurthia</i> sp. [112]	Malachite Green
	<i>Pseudomonas putida</i> [113]	
	<i>Pseudomonas putida</i> [112]	
	<i>Aeromonas hydrophila</i> [114]	
	<i>Micrococcus lylae</i> [115]	Crystal Violet
	<i>Bacillus pumilus</i> [115]	
Fungi	<i>Pseudomonas aeruginosa</i> , <i>Proteus vulgari</i> [116]	
	<i>Aeromonas hydrophila</i> [114]	Safranin-O
	<i>Phanaerochaete chrysosporium</i> [117]	Orange II
	<i>Aspergillus fumigate</i> [118]	Methylene Blue
	<i>Ischnoderma resinosum</i> [119]	Malachite Green
	<i>Aspergillus flavus</i> , <i>Alternaria solani</i> [120]	
	<i>Penicillium janthinellum</i> PI [121]	Crystal Violet
	<i>Ischnoderma resinosum</i> [119]	

	<i>Aspergillus niger</i> [122, 123]	Congo Red, Safranin-O
Algae	<i>Chlorella vulgaris</i> , <i>Anabaena oryzae</i> , <i>Wollea sacata</i> [94]	Orange G
	<i>Synechocystis sp.</i> [124]	Reactive Red
Yeast	<i>Saccharomyces cerevisiae</i> , <i>Saccharomyces uvarum</i> , <i>Saccharomyces lipolytic</i> <i>Turolopsis candida</i> [97]	Reactive Brilliant Red K-2BP
	<i>Saccharomyces cerevisiae</i> MTCC463 [125]	Remazol Black B, Methyl red
	<i>Kluyveromyces marxianus</i> IMB3 [126]	Remazol Black B

Phytoremediation is an alternative green approach to biotreating wastewater contaminated with heavy metals and organic pollutants [21, 127]. The primary benefit of dye removal by plants is that it is an autotrophic system with large biomass, which requires little nutrient cost, is easier to handle, and is generally accepted by the public due to its aesthetic demand and environmental sustainability [128, 129]. Although extensive research has been carried out to establish effective and efficient phytoremediation techniques for decolorizing and degrading azo dyes, large-scale application of phytoremediation is not feasible. Presently it faces several problems, including the extent of pollutants tolerated by the plant and the requirement of large areas to establish treatment plants [130].

The adsorption technique can be traced back to the 17th century when the first qualitative studies on the uptake of gases by charcoal and clays were reported by C.W. Scheele in 1773. It was followed by Lowitz', who studied using charcoal to decolorize tartaric acid solutions. Similar phenomena were observed with vegetable animal charcoals by Larvitz in 1792 and Kehl in 1793, respectively. The term 'adsorption' was proposed by Bois-Reymond and introduced into the literature by Kayser [131]. After that, the adsorption method has been a continuing subject of interest with its wide array of applications for the removal of solutes from solutions and gases from the air atmosphere [4]. Adsorption methods involve the movement of a substance from the bulk liquid to the surface of the adsorbent [4]. The substance that accumulates at the interface is called 'adsorbate,' and the solid on which adsorption occurs is 'adsorbent' [132]. The possible mechanism is due to the interaction between azo dye and adsorbent *via* electrostatic interaction, hydrogen bonding, functional group interaction, π - π interaction, ion exchange, Vander Waals force, hydrophobic interaction, and processes such as surface diffusion and intraparticle pore diffusion as shown in Figure 6 [32, 133].

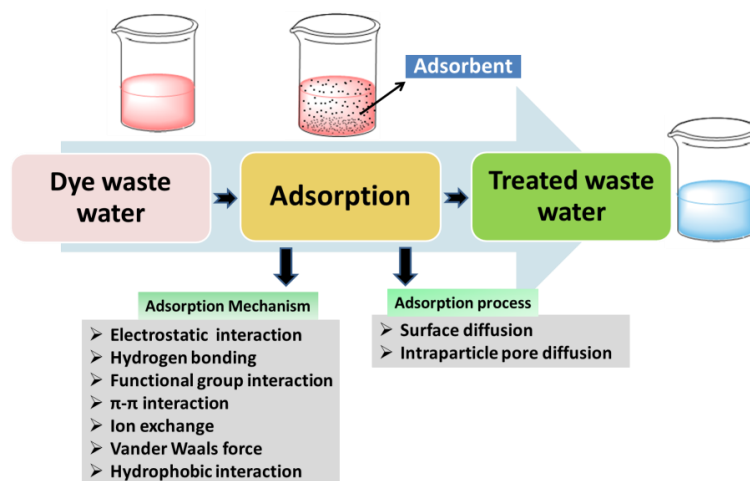


Figure 6 Schematic presentation of the adsorption process.

Several types of adsorbent materials that have been utilized to remove dye molecules effectively from wastewater are metal-organic frameworks [134], magnetic nanocomposite [135], conducting polymers [136], clay minerals, zeolites [137], metal oxides [138], metal hydroxides [139] biopolymers and agricultural wastes [140]. Undoubtedly, a universal adsorbent activated carbon is the most effective adsorbent for the treatment of dye from industrial wastewater [81, 141]. Nevertheless, they are still subjected to considerable disadvantages such as cost, energy consumption, regeneration, and non-selective to all dyes. Therefore, researchers have made an enormous effort and shown keen interest in designing novel adsorbents that are inexpensive, sustainable, biodegradable, renewable, and have good surface adsorption characteristics [142]. The main advantage of the adsorption process is the non-generation of toxic secondary pollutants compared to other methods, which involve toxic chemicals, high energy consumption, and a highly expensive process [143].

Recently, the major focus has been shifted to natural adsorbents than synthetic ones since they are abundant in nature, low in cost, and require little processing. Some of the promising low-cost adsorbents are microorganisms [144], agricultural waste [140], and naturally occurring industrial waste, which is listed in Figure 7 [145]. The application of these low-cost adsorbents has benefited both water treatment and waste management simultaneously.

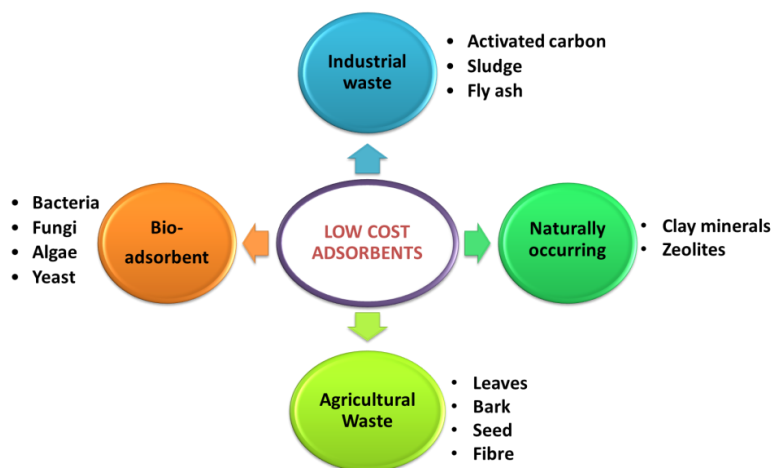


Figure 7 Low-cost adsorbents.

Along with these, several nanomaterials have been widely explored for the adsorption of waste from textile effluent [146, 147]. Carbon nanotubes, oxidized graphene, reduced graphene, boron nitride nanosheets, boron carbon nanosheets, etc., have been widely explored as effective candidates for the adsorption of dyes from wastewater [148-150].

Along with dyes, various metal ions like Cr (VI), Cd (II), Pb (II), and Zn (II), etc., are also present in textile wastewater and pose a serious health hazard; therefore, their removal is essential before discharge into water. The source of heavy metals in the textile effluent is various pretreatment, dyeing, and finishing steps. In addition, some dyes (like chrome acid dye, reactive and direct metal-containing dye) need metal ions for their application and become an important source of metal ions in textile wastewater [151]. Various physical and chemical methods like adsorption [152, 153], coagulation-flocculation [154, 155], ion exchange [155, 156], membrane separation, [157] biological treatment [158], etc. can be used for the removal of metal ions. Each method has its advantages and disadvantages, and the selection of a particular method depends on the type of metal ions and their concentration.

2. Conclusions

The water discharged by the textile industry is highly polluting and contains a variety of toxic metals, chemicals, and dyes. These are hazardous for the aquatic ecosystem and cause various health problems in aquatic animals and human beings. Therefore, textile effluent should be treated before discharging into a water body. Various physical, chemical, and biological methods for treating textile effluent were discussed. All these treatment processes have pros and cons; therefore, selecting the most suitable method can be made based on operational costs and sustainability. The physical and chemical methods are highly expensive, require specialized instruments and other accessories to operate, and are high in energy consumption. The chemical methods require various other chemicals to treat the chemical waste generated by the textile industry. These methods also generate by-products like solid waste and sludge formation, thereby generating a large amount of solid waste. Biosorption offers many advantages due to its simplicity in application, high removal efficiency, and cost-effectiveness. The adsorption method is another convenient process owing to its economic feasibility, simplicity of operation, recycling of absorbents, sludge-free, and high performance. Biological methods use microorganisms and their enzymes to decompose waste material generated by the textile industry. These methods are cost-effective and eco-friendly, but the efficiency of waste material removal is less than physical and chemical methods. Therefore, there is still scope for future development of innovative technologies to treat colored wastewater generated from textile dyeing industries.

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Author Contributions

Both authors contributed equally to this work.

Competing Interests

The authors have declared that no competing interests exist.

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