

Review Article

Phycoremediation of Synthetic Dyes: An Effective and Eco-Friendly Algal Technology for the Dye Abatement

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Rapid industrialization leads to serious environmental hazards due to the increase in the release of pollutants into the environment. Industries that use synthetic dyes for different applications are a predominant source for dye contaminants by releasing the dye in wastewater with pretreatment or without treatment directly into the water bodies, making serious water pollution in the environment. Therefore, it is imperative to safeguard the environment from such contaminants and their associated negative impacts. The conventional treatment method that is used to treat dye-contaminated wastewater is generally costly and has a possibility to produce secondary metabolites. Due to the above problems, the biological method is preferable to treat effluent or dye-contaminated wastewater. Phycoremediation is an algae-based eco-friendly dye abatement technique from contaminated environments. This review highlights the phycoremediation of dyes and its underlying mechanisms along with the information on synthetic dyes, classification, hazardous effects, and other major techniques of dye abatement. This review provides a comprehensive insight into several influencing factors such as pH, temperature, contact time, the dose of algae biomass, and agitation speed, as well as functional groups involved in the phycoremediation process.

1. Introduction

Dyes are becoming more widely used as a result of their numerous applications in various industries. Dyes are difficult to remove from wastewater due to the aromatic molecular structure that makes them much more stable to light, heat, and oxidizing agents [1, 2]. A dye is a substance that is used to impart color to a product or material, i.e., paper, leather, textile, rubber, plastic, and cosmetics [3, 4]. Color added to the product or material creates an attractive appearance and gives added value to an aesthetic for a social reason. Coloring compounds can be made from a natural compound or human made (synthetic). Synthetic dyes are aromatic, water-soluble, and dispersible organic colorants [4]. The use of synthetic dyes significantly increases by the industry such as textile, tannery, paper, printing and dyeing, food, and cosmetics for coloring materials. Dyes help to make the world more beautiful by colored substances; however, they cause serious environmental pollution. In the

world, annually, around one million tons of dyes are produced [5, 6]. A huge amount of synthetic dyes is released into the environment through the textile, pharmaceutical, and chemical industries. From the above three industries, the textile industry is the largest consumer of synthetic dyes.

Synthetic dyes are becoming more common in the textile and dyeing industries due to their ease and cost effectiveness in synthesis, as well as other advantages such as high light stability, firmness against temperature, detergents, and microbes, and color variety, when compared to natural dyes.

During the dyeing process, the main environmental problem associated with the use of dyes is their discharge because the fixation efficiency varies from 60 to 90% [7]. It is also estimated that about 10–15% of the dyes are discharged into wastewater during the dyeing process [8]. As a result, it leads to potential pollution in the environment [9]. Many physical and chemical methods have been applied to solve these issues such as adsorption, chemical precipitation, photolysis, chemical oxidation and reduction,

electrochemical treatment, filtration, coagulation, and chemical flocculation [10–13]. However, these techniques have some disadvantages such as they are expensive, have less decolorization efficiency especially on the synthetic dyes, and are tedious extraction procedures for their organic metabolites. More secondary sludge is generated from these techniques that can cause additional pollution problems as secondary environmental contaminants. Due to these limitations, the bioremediation approach is receiving much attention as the best alternative to physicochemical treatment. Bioremediation is an innovative technology for the treatment of contaminants in which microbes (bacteria, yeast, fungi, and algae) and plants are used to facilitate the degradation or transformation into less harmful types of different toxic chemicals. Phycoremediation is one of the suitable approaches to remove the synthetic dye and maintain the ecosystem using algae, and it has several advantages, including environmentally friendly, high adsorption capacity, quick operation, widely available resources, and low cost.

2. Overview of Dye

2.1. Dye and Its Major Classifications. A dye is a coloring material that is used to provide color or to modify existing color for various substances. Dyes have chromophores and auxochromes that are substantively responsible for their color [14, 15]. Dye is an organic compound that is bright, ionizing, and aromatic. It is usually used in an aqueous solution and displays an affinity for the substrate on which it is applied [16, 17]. Dyes are categorized as either natural or synthetic depending on their source and their ability to color other substances [18]. Synthetic dyes are recalcitrant compounds that comprise the main residue present in the textile dyeing waste [19]. They are complex aromatic compounds formed by chemical synthesis that provide a wide range of colors [20]. Synthetic dyes are grouped into cationic, non-ionic, and anionic types. Under anionic dyes, most demanding water-soluble acids and reactive dyes are shielded. Nonionic dyes, on the contrary, refer to dispersed dyes that are not ionized in an aqueous medium and are listed in Table 1. Among others, anthraquinone-based dyes are the hardest to degrade due to their attached aromatic ring structure.

Color of the dye is a combination effect of chromophores and auxochromes. Chromophores describe the delocalized electron system with double bonds conjugated alternatively; auxochromes refer to electron-withdrawing or electron-donating substituents that enhance color of the chromophores by changing the electron system's overall energy. Some of the essential chromophores include classes of N, CO, NO₂, and quinoids and NH₃, OH, SO₃H, and CO₂H. The bathochromic effect is shifting adsorption bands to longer wavelengths on a conjugated system of the dye that is advanced by the combination of both chromophores and auxochromes. Also, instead of increasing the chromophores in color processing, auxochromes are responsible for dye solubility and increasing its reactivity to fibers [9]. In anionic and nonionic dyes, the chromophores are mostly azo classes.

During reductive cleavage, the azo group produces toxic amines. Azo-based chromophores contain reactive dyes such as vinyl sulfone, chlorotriazine, trichloropyrimidine, and difluorochloropyrimidine [9].

2.2. Classification of Synthetic Dyes. Dyes are broadly categorized based on (i) chromophoric groups in their chemical structures as azo dyes, anthraquinone dyes, phthalocyanine dyes, etc., (ii) their method of use or application as polyester dyes and cotton reactive dyes, and (iii) their dissociation into an aqueous solution as acidic, basic (cationic), direct reactive (anionic), and disperse/nonionic [21, 22].

Reactive dyes are the only textile dyes designed during the application process to form a covalent bond with the substrate. The reactive dyes offer a wide range of shades of good light fastness and excellent cotton wash fastness. These properties situate this class of dyes at the end of the market quality [23], but massively used by the textile industry. The three most common classes of reactive dyes are azo, anthraquinone, and phthalocyanine [24]. Due to their color intensity and high cellulose reactivity, reactive azo dyes are the most important group of dyes used in the textile industry [25].

Acid dyes are used to color nylon, wool, silk, leather, paper, food, and cosmetics. Acid dyes include chemical groups of azo, anthraquinone, triphenylmethane, azine, xanthene, nitro, and nitroso [26]. Acid dyes are generally bright with a variable stronghold to washing. These dye molecules have great structural variance and include some metal complexes [23].

Azo dyes are the largest group of dyes with a functional group of N=N as a chromophore in an aromatic system. Chromophores contain functional groups such as -N=N-, -C=O, -NO₂, and O=C₆H₄=O (quinoid assemblies), and auxochromes contain functional groups such as -NH₃, -COOH, -OH, and -SO₃H [27, 28]. Azo groups are classified into monazo, disazo, trisazo, tetrakisazo, and polyazo dyes based on the number of azo groups [29].

Triarylmethane dyes are hydrocarbon derivatives that contain two types such as acidic and basic. Triarylmethane acid dyes contain at least two groups of SO₃H which are used to indicate wool and silk dye fibers. Besides, dyes that comprise only one group of SO₃H are used as indicators, such as phenolphthalein. On the contrary, basic triarylmethane dyes are massively used in the production of stamping inks, writing, and printing [30].

The sulfonic acid group in anthraquinone dyes is responsible for their water solubility. Mainly, these groups of dyes are used to dye in wool and silk industries due to their ability towards auxiliary binding agents. Food coloring dyes are a subclass of acid dyes that are used to dye protein fibers and certain nylon fibers at high temperatures [31].

Direct dyes are massively used to dye protein fibers and dye synthetic fibers such as nylon and rayon. These dyes lack fast-drying features after applied to fabrics. Basic dyes formed a colored cationic salt when dissolved in water. These cationic salts bind with an anionic substrate. Basic dyes are also known as cationic dyes and are found to be powerful coloring agents for acrylic fibers [8].

TABLE 1: Characteristics of synthetic dyes [9, 15, 23].

Synthetic dyes	General description	Uses	Ionic nature	Light fastness	Washing fastness	Hot pressing fastness	Dry cleaning fastness	Seawater fastness	Solubility	Application pH
Acid dye	Originated from basic dye acidification; complete color range	Primarily for wool and silk; also, acetate, acrylic, and nylon	Anionic	Very good	Poor	Not affected	Good	Fair	Soluble in water	4-5
Basic dye	First synthetic dye (1856), first coal tar dye	Primarily for wool, silk, nylon, and cotton	Cationic	Poor	Poor	Not affected	Mostly poor	Very poor	Soluble	5-6
Direct dye	Dyes cellulosic fabrics directly; some dye wool and silk	Primarily for cellulosic fabrics	Anionic	Good to excellent	Poor	Good	Good	Poor to good	Depends on the types of direct dyes	7
Disperse dye	Developed for acetate	Primarily for acetate, also, polyester, nylon, and cellulose fibers	Nonionic	Fair to excellent	Fair to good	Some color change is possible	Good	Good	Slightly soluble in water	4-5
Reactive dye	Forms a covalent bond with the fiber	Primarily for cotton apparel	Anionic	Very good	Good	Not affected	Good	Good	Depends on the types of reactive dyes	11-13
Sulfur dye	Insoluble in water, complete shade range	Used for linen, cotton, and jute	Nonionic	Poor to fair	Poor to good	Good	Good	Good	Insoluble	10-11
Vat dye	Synthetic indigo original	Used for cotton and wool	Nonionic	Excellent	Good	Good	Good	Good	Insoluble, soluble leuco salts	12-13

Disperse dyes are water-insoluble nonionic dyes that are commonly used on polyester, nylon, cellulose, and acrylic fibers. They encompass several groups such as azo, anthraquinone, styryl, nitro, and benzodifuranone. The dyes used for plastics, gasoline, lubricants, oils, and waxes are solvent soluble. These dyes are principally azo and anthraquinone chemical classes. The dyes used for coloring cotton, rayon, silk, leather, paper, and wood are called sulfur dyes. Vat dyes are insoluble in water and used to dye cellulosic fibers. The main chemical groups of these dyes are anthraquinone and indigoids [8].

2.3. Hazardous Effects of Synthetic Dyes. The effluent discharged from the dyeing industries is the major cause of water pollution because it contains a huge volume of dyes. The high concentration of dyes in water bodies blocks sunlight and reduces the oxygenation potential of receiving water, affecting aquatic biodiversity and photosynthesis. Blue, green, or brown color of watercourses is accepted somehow by the public, but red and purple colors in water bodies receive people's concern [32]. The natural pigment used for coloring textile fades easily when it is exposed to

sunlight, heat, and water. Approximately 15% of textile dyes are lost and released into water during the dyeing process. This effluent eventually triggers issues such as mutagenic, carcinogenic, and toxic effects [8]. Synthetic dyes pose widespread health hazards by their nature or the nondegradable byproducts which are produced with the reaction of other chemicals [28].

The industrial effluents containing dyes affect the growth of microorganisms and photosynthetic activities of aquatic flora by reducing light penetration in receiving water bodies. Moreover, discharged dyes reduce the amount of dissolved oxygen in water by forming a thin layer on the surface of the receiving water body. Furthermore, this kind of effluent increases the chemical oxygen demand (COD) which is an indication of high-level pollution [33]. Many of the dyes are carcinogenic and pose a serious hazard to aquatic living organisms such as damage to the liver, gills, intestine, and gonads and destroy aquatic communities in the ecosystem (Figure 1) [35]. Direct use of untreated dyeing effluents for irrigation has serious environmental consequences, such as reduced soil fertility, seed germination, chlorophyll, and protein content in plants [8].

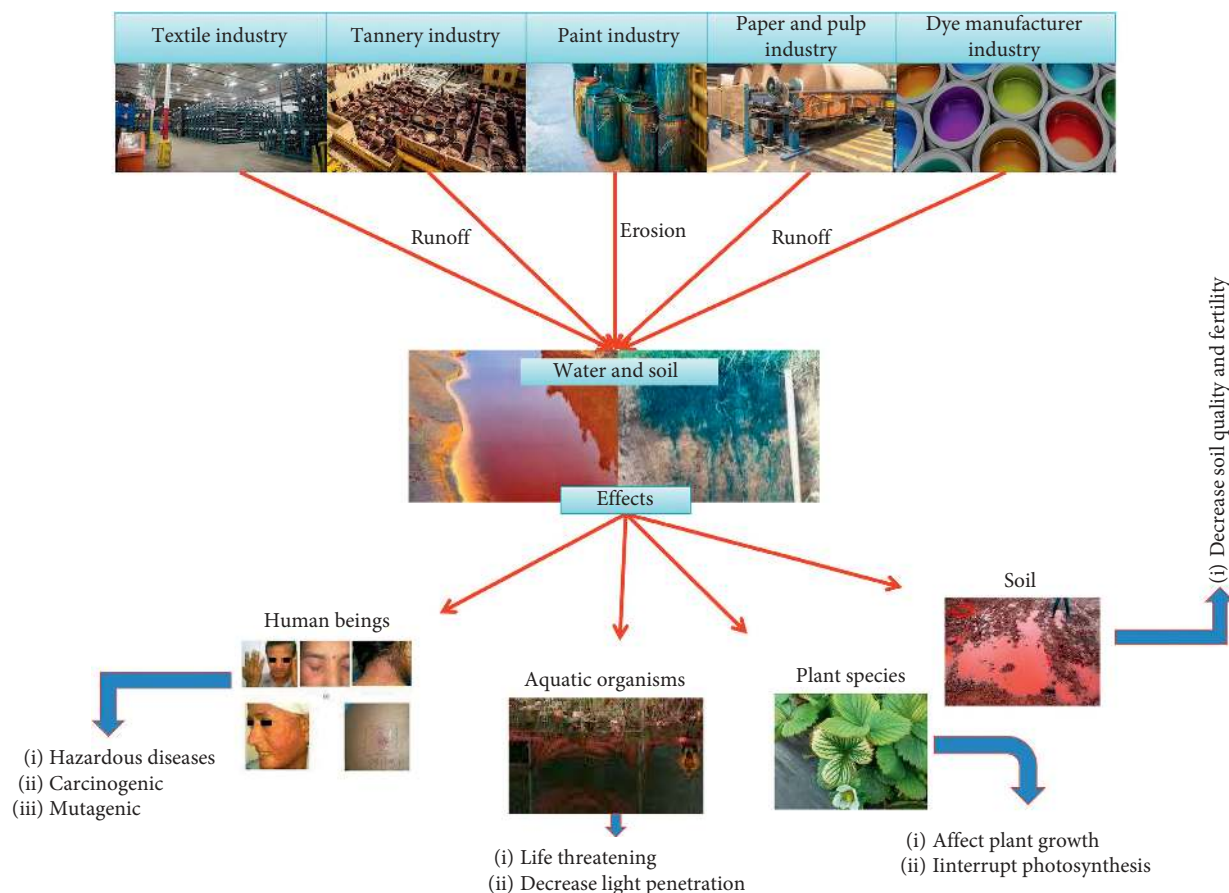


FIGURE 1: The hazardous effects of synthetic dyes on the environment from different industries [34].

Azo dyes can cause multiple health issues to humans such as skin irritation, chemosis, contact dermatitis, exophthalmos, lacrimation, rhabdomyolysis, permanent blindness, vomiting gastritis, acute tubular necrosis supervene, hypertension, vertigo, and, upon ingestion, edema of the neck, face, tongue, pharynx, and larynx along with respiratory distress [6]. Some dyes cause bladder cancer, splenic sarcomas, hepatocarcinoma, and nuclear abnormalities; in laboratory animals, some dyes cause splenic sarcomas, hepatocarcinoma, and nuclear abnormalities [36]. Through the chemical process, the dyes undertake alteration in the chemical structure resulting in the formation of new xenobiotic compounds that have a great impact on the environment. Additionally, they can also cause skin irritation, allergy, cancer, neurobehavioral disorders, autoimmune diseases, and other human health disorders such as nausea, vomiting, and paralysis and cause severe damages to the liver, kidney, reproductive system, brain, and central nervous system [21, 37, 38]. Some dyes with monoazo or anthraquinone structures have been found to cause hypersensitivity reactions, such as skin irritation or dermatitis [39].

3. Methods for Synthetic Dye Abatement

Different conventional technologies such as chemical, physicochemical, and biological methods which are used to treat wastewater contaminated with dyes are summarized in Table 2.

3.1. Conventional Technologies. Conventionally, several physical and chemical methods including ultrafiltration, adsorption, flocculation, coagulation, ozonation, advanced oxidation processes, photocatalytic oxidation, Fenton process, and chemical and electrochemical coagulation were applied for the removal of color [22]. All the aforementioned methods have limitations such as incomplete removal of dyes from the environments, sludge production, short half-life (20 min) of O_3 , formation of byproducts, the release of aromatic amines, high cost of electricity, very expensive, ineffective against dispersing and vat dyes, and the repetitive or frequent usage of chemicals leads to secondary pollution problems [9, 40]. Hence, an alternative technique is needed for the abatement of synthetic dyes.

TABLE 2: Methods of synthetic dye biodegradation and decolorization [32, 41, 55].

<i>Physical method</i>	
Adsorption	Refers to a process where a substance or material is concentrated at a solid surface from its liquid or the gaseous surrounding.
Irradiation	Involves the use of radiations usually obtained from monochromatic UV lamps working under 253.7 nm. It is a simple and effective technique for removing a wide variety of organic contaminants and disinfecting harmful microorganisms.
Filtration processes	Microfiltration: it is mostly employed for the treatment of dye baths containing pigment dyes.
	Ultrafiltration: this technique can remove polluting substances such as dyes only 31–76% but can be used to eliminate macromolecules and particles.
Reverse osmosis	Nanofiltration: it is employed for the treatment of colored effluents from the textile industry mostly in a combination of adsorption and nanofiltration as NF modules are very sensitive to fouling by colloidal materials and macromolecules. This technique is used to eliminate hydrolyzed reactive dyes, most types of ionic compounds, and chemical auxiliaries in a single step.
<i>Chemical method</i>	
Electrochemical	This process is very simple and is based on applying an electric current to wastewater by using sacrificial iron electrodes to produce ferrous hydroxide.
Oxidative processes	Oxidation by ozonation is a technology initially used in the 1970s, and it is carried out by ozone generated from oxygen.
	Chemical oxidation is the conversion of pollutants by chemical oxidizing agents (such as chlorine, ozone, Fenton reagents, UV/peroxide, and UV/ozone).
	Oxidation with sodium hypochlorite: in this treatment, azo bond cleavage is initiated and accelerated by the attack of the dye molecule by Cl^+ at the amino group.
	The oxidation processes with hydrogen peroxide (H_2O_2) can be used as wastewater treatment in two systems: (1) homogenous systems based on using visible or ultraviolet light, soluble catalysts such as Fenton reagents which are strong oxidants compared to H_2O_2 and other chemical activators such as ozone and peroxidase. (2) Heterogenous systems based on using semiconductors, zeolites, and clays with or without ultraviolet light.
Photochemical oxidation: the UV treatment of dye-containing wastewater in the presence of H_2O_2 can break down the dye molecules into smaller organic molecules or even ultimate products such as CO_2 and H_2O and other inorganic oxides.	
Coagulation	Coagulation of dyes and other auxiliaries in textile effluents has been successfully done by aluminum, iron slats, organic polymer, flocculants, etc.
Electrocoagulation	Electrocoagulation is an advanced electrochemical treatment for dye and color removal. It involves processes such as electrolytic reactions at electrodes, coagulation in the aqueous effluent and adsorption of soluble pollutants on coagulants, and, finally, their removal by sedimentation
<i>Biological method</i>	
Bacterial	Aerobic biological treatment: use bacteria and oxygen (from injected air) to remove dissolved organic load (COD/BOD) from dye-containing wastewater. The process is controlled by oxygen sensors in the activated sludge (aeration tank), and residual bacteria (waste activated sludge) can be separated in various ways.
	Anaerobic biological treatment: anaerobic biodegradation of water-soluble dyes including azo dyes is mainly reported to take place by a redox reaction with hydrogen leading to the formation of methane, carbon dioxide, hydrogen sulfide, and other gaseous compounds and releasing electrons.
Fungal	Fungal organisms can decolorize a wide range of dyes.
Microalgal	Algae are capable of decolorizing colored wastewater through mechanisms of enzymatic pathways as well as adsorption on algal biomass.
Enzymatic	The enzyme that is produced from microbes and plants is used to degrade or decolorize dyes.

3.2. Biological Treatment Methods. The biological method (bioremediation) is considered the best alternative over conventional physicochemical treatment due to its potential advantages of being inexpensive and nonhazardous. Bioremediation (using microbes) is a pollution control technology where the biological systems are used to drive the degradation or transformation of various toxic chemicals into less harmful forms. Biological treatment methods are eco-friendly methods that are gaining importance in today's scenario. Microorganisms such as bacteria, fungi, algae, yeast, and their enzymes can be successfully utilized to remove color of a wide range of dyes through anaerobic, aerobic, sequential anaerobic-aerobic treatment, and bioadsorption processes [41–43].

3.3. Phycoremediation. Phycoremediation means the use of macroalgae, microalgae, and cyanobacteria for the removal or biotransformation of contaminants, containing nutrients, synthetic dyes, heavy metals, and xenobiotics from dye-contaminated wastewater and CO_2 from waste air (for environmental cleanup) [21, 44]. Removal of nutrients from wastewater, acidic and metal wastewater treatment, degradation, transformation of recalcitrant compounds, and toxic compound detection by using algal-based biosensors are some of the key applications of phycoremediation [45]. Besides, abatement of synthetic dyes by algal biomass possesses advantages such as eco-friendly, high adsorption efficiency, fast process, widely available resources, and low cost [46].

4. Algae

Algae are derived from the Greek word “alga” which means “phyco.” These organisms are heterogeneous, predominantly eukaryotic, and aquatic organisms that differ from microscopic cells to highly differentiated plants. Algae are categorized as either aquatic or freshwater plants which have very high carbon trapping and photosynthetic efficiencies when compared to terrestrial plants [47]. Algae are classified as “thallophytes” or plants that lack roots, stems, and leaves. Chlorophyll is the dominant photosynthetic pigment in these species. On the contrary, they lack a sterile covering of cells around the reproductive cells. The green algae are dispersed in many regions of the world, from being airborne or subaerial to being terrestrial or aquatic, either in freshwater or marine habitats [48]. Algae include macroalgae and microalgae, and many are eukaryotic organisms. Due to this, they have the properties such as those with cells displaying a high degree of internal organization, including a membrane-bound nucleus containing the genetic material and several other internal parts, organelles that are also surrounded by membranes. On the contrary, the term also consists of cyanobacteria or blue-green algae, which are prokaryotic (those cells that lack a distinct nucleus) [47]. Algae can play an important role in the removal of azo dyes and aromatic amines in stabilization ponds [49]. The dye abatement efficiency of algae is varied based on the dyes’ molecular structure and the algae species used for the process [50]. Algae accumulate colored water during their growth and potentially decolorize dye-contaminated wastewater. The resulting biomass is being processed further for the production of bioenergy and algal-based bioproducts [51]. The selected algae used for the abatement of various synthetic dyes are listed in Table 3.

4.1. Microalgae. As the name specifies, these are microscopic algae and can be motile or nonmotile depending on the presence of the flagella. Microalgae are unicellular and photosynthetic microorganisms, usually in the size range of 1–400 μm , and visible by using a microscope [37, 75]. In terms of biomass, microalgae are one of the largest primary producers and responsible for at least 32% of global photosynthesis and oxygen production as well as the best known CO_2 sequestration [76, 77].

4.2. Macroalgae. Macroalgae are multicellular organisms and are generally seen without the aid of a microscope. Seaweeds, also known as macroalgae, are macroscopic multicellular algae with defined tissues and specialized cells. Macroalgae have cell types that are similar to those of terrestrial vascular plants. Furthermore, the macroalgae may be either marine or freshwater [47, 78]. Macroalgae can accumulate heavy metals, synthetic dyes, and other contaminants reaching concentrations that are thousands of times higher than the corresponding concentrations in seawater. Furthermore, algae fulfill the criteria for bio-indicators as they are widely distributed. The microalgae have unique features such as suitable size for high

accumulation of metals, easy to identify, and collection and recovery are simple. Macroalgae are grouped into three such as red (Rhodophyta), brown (Phaeophyceae), and green (Chlorophyta) [79]. Macroalgae composition differs based on the species variety. In red algae, galactose-based polyglucans such as agar and carrageenan are majorly observed. The kind of algae that encompass alginate and a large amount of laminarin is called brown algae, whereas green algae principally contain starch and cellulose [80, 81].

5. Factors Affecting the Abatement of Synthetic Dyes by Algae

The molecular structure and microbial activity are influenced by the culture condition that is necessary for effective abatement of dyes. The algae efficiency of biosorption can be affected by optimizing the operating conditions such as pH, temperature, biosorbent dosage, and agitation [82].

5.1. Effect of pH. pH is one of the most important parameters that affects the biosorption efficiency of algae [83]. Besides this, it also affects color and the solubility of some dyes. The interface between sorbate and biosorbent is affected by pH of an aqueous solution. The biosorbent surface has numerous functional groups such as carboxyl, hydroxyl, amino, and phosphates. As a consequence of this, the net charge of biosorbent is dependent on pH. If pH decreased, the biosorbent surface had more positively charged sites, which were favorable to the adsorption of dye anions due to the electrostatic attraction [84]. At lower and higher pH, the biosorbent surface becomes net positive charge and net negative charge, respectively. Therefore, at lower pH, the biosorbent surface binds with anionic dyes, and also at higher pH, the biosorbent surface is attached with cationic dyes [85]. Aravindhana et al. [86] observed that uptake of Sandocryl Golden Yellow C-2G by green seaweed *Caulerpa scalpelliformis* increases from 17 to 27 mg/g with an increase in pH from 3.0 to 8.0. Initial pH of the solution appreciably influences the biosorption of dyes due to a change in the surface properties of the adsorbent [87].

5.2. Effect of Temperature. Temperature plays a great role in the biodegradation of dyes. The viscosity of the solution containing the dyes decreased with an increase in temperature. The amount of adsorbed dye decreased with increasing temperature, indicating the exothermic nature of the biosorption process [70]. The decrease in the efficiency of the adsorption due to temperature increases can be dedicated to changes in active sites of the adsorbent and also the tendency of the adsorbed material to get away from the active sites into the aqueous solution [72]. Aravindhana et al.’s results [86] illustrated that the sorption capacity uptake of Sandocryl Golden Yellow C-2G by green seaweed *Caulerpa scalpelliformis* decreased from 28 to 23 mg/g with a temperature increase from 20 to 60°C.

TABLE 3: Abatement of various synthetic dyes by algae.

Algae	Synthetic dyes	Experimental conditions: initial dye conc. (mg/L), pH, time (hr), temp (°C), biomass (g/L)	% removal	References
<i>Chlorella vulgaris</i>	Aniline blue	25, —, —, 264, —, —	58	[52]
<i>Gonium</i> sp.	Reactive Blue 220	—, 8, 336, —, —	84.2	[53]
<i>Gracilaria verrucosa</i>	Phenoxyalkanoic acid herbicide 2,4-D	—, —, —, —, —	47	[54]
<i>Haematococcus</i> sp.	Malachite green	100, —, —, —, 1	67	[55]
<i>Oscillatoria</i> sp.	Blue dye	—, 10, 336, —, 3	76.48	[1]
	Red dye	—, 10, 336, —, 3	62.63	
<i>Spirogyra</i> sp.	Blue dye	—, 10, 336, —, 3	78.29	[1]
	Red dye	—, 10, 336, —, 3	64.21	
<i>Scenedesmus bijuga</i>	Monoazo and diazo	—, —, 48, 25, —	68	[31]
<i>Vaucheria</i> sp.	Malachite green	—, 8.5, 17.5, —, —	85.9	[25]
<i>Coelastrella</i> sp.	Rhodamine B	100, 8, 480, 30, —	80	[56]
<i>Chlorella vulgaris</i>	Methylene blue	100, —, —, —, —	83.04	[57]
<i>Ulva lactuca</i>	Methylene blue	25, 10, —, —, 2.5	4.012	[58]
<i>Sargassum</i> sp.	Acid Black 1	40, 4.2, 0.5, —, —	96.8	[59]
<i>Oedogonium subplagiostomum</i> API	Methyl orange	500, 6.5, 132, —, 0.4	97	[60]
<i>Sargassum crassifolium</i>	Malachite green	5, 8, 2.5, 25, 2	98.3	[61]
<i>Chlorella vulgaris</i>	Yellow dye	10, —, 336, —, —	3.12	[17]
<i>Sphaerocystis Schroeteri</i>	Yellow dye	10, —, 336, —, —	45.03	
<i>Cosmarium</i> sp.	Malachite green	10, 9, —, —, —	87.1	[62]
<i>Desmodesmus</i> sp.	Methylene blue		98.6	[63]
	Malachite green	20, —, 144, —, —		
<i>Spirogyra</i> sp. (CKW1) and <i>Cladophora</i> sp. (PKS33)	Reactive blue azo dyes	100, 7, 168, 30, —		[64]
<i>Spirulina platensis</i>	Acid Black 210	125, 2, 1, 60, 0.5	98.55	[65]
	Acid Blue 7	125, 2, 1.25, 60, 0.5	97.05	
	Reactive Black 5	200, 5, 240, 40, —	80	
<i>Chlorella vulgaris</i>	Direct Blue 71	200, 8, 240, 40, —	78	[66]
	Disperse Red 1	300, 8, 240, 40, —	84	
	Malachite green	5, —, 120, —, 0.2	93	
<i>Oscillatoria</i> sp.	Methylene blue	5, —, 120, —, 0.2	66	[67]
	Safranin	5, —, 120, —, 0.2	52	
	Malachite green	100, 5, —, 50, —	97.13	
<i>Nostoc</i> sp.	Malachite green	100, 5, —, 50, —	97.13	[54]
<i>Ulva lactuca</i>	Methylene blue		67	[35]
	Malachite green	100, 6, 1, —, 0.1	78	
<i>Ulva lactuca</i>	Methylene blue	25, 8, 2.83, —, 1.25	91.92	[4]
<i>Spirogyra</i> sp.	Synazol	—, 3, 18, 30, 8	85	[68]
<i>Chlorella</i> sp.	Malachite green	—, 10, 2.5, 25, -	80.7	[69]
<i>Ulothrix</i> sp.	Methylene blue	90, 7.9, 0.5, 37, 1	86.1	[70]
<i>Turbinaria conoides</i>	Acid Blue 9	100, 1, 3.75, 33, 3	87.64	[71]
<i>Padina sanctae-crucis</i>	Methyl violet	10, 8, 1.33, 25, 2	98.85	[72]
<i>Chara</i> sp.	Malachite green	9.7, 6.8, 1.25, —, 3.9	57.81	[25]
<i>Scenedesmus quadricauda</i>	Malachite green	6, 6, 1.15, —, 0.004	73.49	
<i>Chlorella vulgaris</i>	Malachite green	6, 6, 1.15, —, 0.004	91.61	[73]
<i>Chlorella vulgaris</i>	Congo red	50, —, 216, 35, —	100	[74]

5.3. *Effect of Contact Time.* The dye uptake rate of sorbent species is rapid at the beginning of the contact time; however, as the contact time reaches the equilibrium, the uptake rate is reduced or halted. For instance, in the study of Pratiwi et al. [4], the adsorption efficiency increased with a rise in contact time up to 110 min and later became less constant. There are a massive number of vacant surface sites which are accessible for sorption during the initial stage. Subsequently, the vacant surface sites are occupied, and it

becomes hard to bind easily as a result of repulsive forces between the solute molecules on the solid and bulk phases [8].

5.4. *Effect of Initial Concentration of Dyes.* Dye concentration has a major influence on the dye abatement process. The adsorption efficiency is directly proportional to the pollutant concentration. Hence, the efficiency is generally found to be

higher in lower dye concentration and subsequently reduced with an increment in dye concentration [88]. It is obvious that, as the adsorbent dose increases, the number of available adsorption sites increases; this, in turn, increases the uptake of the dye. For instance, according to Aravindhana et al. [86], the amount of uptake of Sandocryl Golden Yellow C-2G adsorbed by macroalga *Caulerpa scalpelliformis* increases from 4.86 to 33.15 mg/g for an increase in initial dye concentration from 25 to 200 mg/L. Al Hamadi et al. [65] reported that the adsorption capacity of Acid Black 210 and Acid Blue 7 by *Spirulina platensis* was increased from 94.37% to 97.7% and 87.5% to 91.6%, respectively, by increasing the initial dye concentration from 25 to 125 mg/L.

5.5. Effect of Agitation Speed. Agitation is crucial for equal mixing of the medium components, dispersion of cells, and nutrients, as well as mass transfer phenomena in the dye abatement process. In addition to this, agitation also leads to shear force, which affects microbes in various dimensions that include morphological changes, rupture of the cell wall, variation in their growth, and product formation [8]. According to Dođar et al. [70], the increase of the mixing rate improves the transport of dye molecules from the bulk to the surface of the cell (film/boundary layer diffusion). In the study of Jerold and Sivasubramanian [89], an increment of agitation rate from 50 to 150 rpm increased the uptake rate of malachite green from 21.5 to 56.2 mg/g by macroalga *Sargassum swartzii*.

5.6. Effect of Biomass Dosage. The dosage of the biosorbent (biomass concentration) is also another factor that influences the dye abatement capacity of biosorbents. There are direct relationships between biomass concentration and biosorption, which mean the number of biosorption sites increases with an increased dosage of biomass concentration that leads to efficient biosorption of dyes [28, 90]. The study of Aravindhana et al. [86] reported that the amount of basic blue dye adsorbed by *Caulerpa scalpelliformis* was decreased from 54.16 to 19.58 mg/g with an increase in the concentration of seaweed biomass from 0.15 to 0.5 g/50 mL. According to Kumar et al. [91], cationic dye uptake by *Pithophora* sp. was decreased from 42.2 mg/g to 9.2 mg/g (57.8% to 90.8%) with an increase in biomass concentration from 0.02 g to 0.06 g. Therefore, it leads to an increment in the solute transfer rate onto the adsorbent surface, which means the amount of solute adsorbed onto the unit weight of adsorbent gets split with increasing biosorbent dosage [92].

6. Mechanism of Dye Abatement by Algae

Different microorganisms may have different pathways for degrading different dyes depending upon the dye structure, strategy of the microbial system for dye degradation, and many other factors. Even small structural differences can affect the decolorization process [85]. The way of microbial removal of dyes is generally adsorption or degradation [93]. Biosorption implies removing the dye molecules from the aqueous phase to the solid phase (the bioadsorbent), while

biodegradation entails the enzymatic breakdown of the chemical bonds that constitute the chemical structure of the dyes [94]. Most studied algae that are involved in the decolorization and degradation of dyes are blue-green algae, green algae, and diatoms [36]. Algae biomass has the highest biosorption efficiency and electrostatic force of desirability for pollutants in wastewater due to its wide surface area and high binding ability. Several studies have shown that metabolites of pollutants found in wastewater are adsorbed by the algae surface [28].

Mechanisms of algal decolorization can involve enzymatic degradation, adsorption, or both. Color removal by algae is mainly due to three intrinsically different mechanisms such as assimilative utilization of chromophores for the production of algal biomass, CO₂ and H₂O transformation of colored molecules to noncolored ones, and adsorption of chromophores on algal biomass (Figure 2) [21, 28, 95, 96]. Algae are capable of degrading azo dyes through an induced form of azoreductase. They promote the reductive cleavage of the azo bonds and release the aromatic amines that will be degraded in CO₂ and H₂O [33, 97].

Algae have a high surface area and high binding affinity during biosorption that facilitate the biosorption capacity. The electrostatic attraction and complexation are known to take place during algal biosorption [94]. The functional groups found on the cell surface of algae are responsible for decolorization by attracting dye contaminants from the environments. The biosorption capacity of algae is mainly determined by the cell wall. The presence of the heteropolysaccharide and lipid compound on the cell wall of algae contains many functional groups such as amino, carboxyl, hydroxyl, and phosphate groups, which are responsible for causing strong attractive force between dyes and the cell wall [98]. Alginate has an important role in adsorption by brown algae through binding to pollutant ions. Generally, metal ions such as K⁺, Na⁺, Ca²⁺, and Mg²⁺, which are available on the algal biomass, are obtained from seawater which is bound to the acid functional groups [99].

7. Functional Group Involvement during Dye Abatement by Algae

Fourier-transform infrared (FTIR) spectroscopy will be used to determine the infrared spectrum of the major functional groups of the microbes. FTIR spectroscopy has the advantages of its simple, inexpensive, fast, noninvasive, and multiplex measurements. FTIR spectroscopy has already been used to study the biosorption of dyes from wastewaters using algae. The selected algal functional groups involved in the decolorization of different synthetic dyes are summarized in Table 4. According to the investigation of Foroutan et al. [100], *Sargassum oligocystum* has around six functional groups or active sites that adsorb (avoid toxicity) methylene blue and methyl violet. Similarly, the findings of Brahmabhatt and Jasrai [1] stated that *Oscillatoria* sp. and *Spirogyra* sp. have many active sites on their cell such as alcohol or phenol, alkynes, primary amines, chloroalkanes, and bromoalkanes for the blue dye decolorization process. Besides, other studies stated that the *Oscillatoria* sp. and *Spirogyra* sp. algal

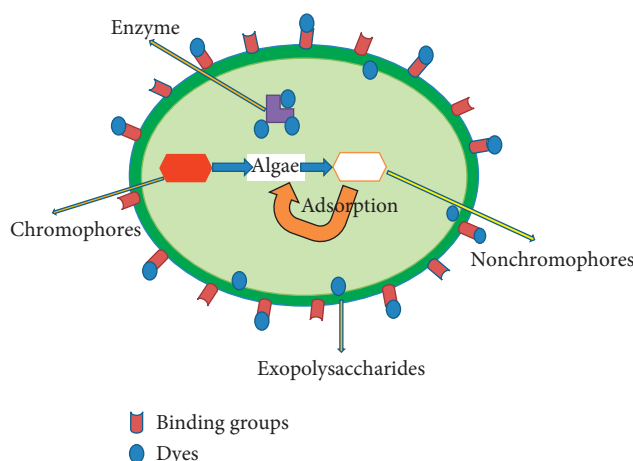


FIGURE 2: Mechanisms of dye adsorption by algae [28, 94].

TABLE 4: Algal functional group involved in decolorization of different synthetic dyes.

Algae	Synthetic dyes	Functional groups involved	References
<i>Sargassum oligocystum</i>	Methylene blue Methyl violet	C=C, C=O, C-H, C-O, Fe-O, and C-C	[100]
<i>Oscillatoria</i> sp. and <i>Spirogyra</i> sp.	Blue dye Red dye	OH, \equiv C-H, N-H, and C-X(X=Cl, Br) OH, \equiv C-H, N-H, C=O, C-H, and C-X (X=Cl, Br)	[1]
<i>Sargassum muticum</i>	Methylene blue	O-H —NH ₂ , C=O, and C-O	[101]
<i>Phormidium animale</i>	Remazol Black B	C-H and CH ₃	[102]
<i>Oedogonium subplagiostomum</i> AP1	Methyl orange	O-H, N-H, and —N=N—	[60]
<i>Spirulina platensis</i>	Reactive Red 120	O-H, NH ₂ , CH ₂ , C-N, P-O, S-O, and C-H	[103]
<i>Spirulina platensis</i>	Tartrazine and allura red	O-H, NH ₂ , CH ₂ , C=C, —S-O, —P-O, and —CH	[104]
<i>Durvillaea antarctica</i>	Methylene blue	O-H, C-H, C=O, —C=C-, and —CH ₃	[105]
<i>Padina gymnospora</i>	Methylene blue	N-H, C-H, R-N=C=S, C=C, and S-S	[106]
<i>Chlamydomonas variabilis</i>	Methylene blue	—OH, —NH, C-H, HC=O, and R ₂ C=O	[107]
<i>Ulva lactuca</i>	Malachite green	—OH, —NH, CH, —COO-, —SO ₃ , and —C-O	[61]
<i>Sargassum crassifolium</i>	Malachite green	—OH, —NH, CH, —COO-, —SO ₃ , —C-O, and —C = O	[61]
<i>Gracilaria corticata</i>	Malachite green	OH, —NH, CH, —SO ₃ , —C-O, and —C = O	[61]
<i>Dictyota cervicornis</i>	Methylene blue	N-H, O-H, —CH, C-CH ₃ , C=O, C-OH, P-O-C, S O, and C-S-O	[99]
<i>Caulerpa scalpelliformis</i>	Sandocryl Golden Yellow C-2G	—OH, —COOH, C=O, and C-O	[86]
<i>Kappaphycus alvarezii</i>	Rhodamine B	N-H, H-O/N, COO ⁻ , C=N, C-O-C, N-O, —OH, C-H, C=O, C=C, C-N, C-O, and C-Cl	[90]
<i>Gracilaria salicornia</i>	Rhodamine B	N-H, H-O/N, COO ⁻ , C=N, C-O-C, N-O, —OH, C-H, C=O, C=C, C-N, C-O, and C-Cl	[90]
<i>Gracilaria edulis</i>	Rhodamine B	N-H, H-O/N, COO ⁻ , C=N, C-O-C, N-O, —OH, C-H, C=O, C=C, C-N, C-O, and C-Cl	[90]
<i>Enteromorpha prolifera</i>	Direct Fast Scarlet 4BS	O-H, N-H, —SO ₃ , C=O, C-N, and PO ₄	[84]
Brown algae	Crystal violet	O-H, N-H, C-H, C=O, C-OH, O-C-O, and —C-O	[108]
<i>Fucus vesiculosus</i>	Methylene blue	O-H, C-H, C=O, N-H, and C-O	[109]
<i>Spirulina maxima</i>	Methylene blue	O-H, C-H, C=O, N-H, and C-O	[109]
<i>Chlorella pyrenoidosa</i>	Methylene blue	O-H, C-H, C=O, N-H, and C-O	[110]
<i>Laminaria japonica</i>	Methylene blue	—OH, —NH, —CH ₂ , —CH ₃ , C=O, C-O, —COOH, and C-O-C	[110]
<i>Spirulina</i> algae	Direct Yellow 12	OH, C=C, C-H, and C=C	[111]
<i>Padina sanctae-crucis</i>	Methyl violet	—OH, —NH ₂ , —C-H, —C=O, C-O, —C-N-C, and S=O	[72]
<i>Chaetophora elegans</i>	Methylene blue	—OH, —NH ₂ , —CH, COOH, >C=O, C-O, >S=O, and C-N	[112]
<i>Chara</i> sp.	Malachite green	C=C, C-N, C-H, —CH ₃ , and —C-N-	[25]
<i>Scenedesmus quadricauda</i>	Malachite green	—OH, —NH, and COOH	[73]
<i>Chlorella vulgaris</i>	Malachite green	—OH, —NH, and COOH	[73]
<i>Sargassum swartzii</i>	Malachite green	NH ₂ , C=O, C-O, C-O, and C-H	[89]
<i>Laminaria japonica</i>	Malachite green	—OH, —NH, C-H, C=O, and C-O	[113]

TABLE 4: Continued.

Algae	Synthetic dyes	Functional groups involved	References
<i>Nostoc comminutum</i>	Remazol Black 5 and Remazol Brilliant Blue	-OH, -NH, -C=O, N-H, -C-O, and -C-N	[114]
<i>Nostoc carneum</i>	Methyl orange	C-H, =C-H, C=O, C=C, NO ₂ , P-OH, S=O, O-H, N-H, P-O, and C-S	[115]
<i>Vaucheria</i> sp.	Malachite green	C=C, C-N, C-H, and -CH ₃	[25]

cell have functional groups that encompass alcohol, phenol, terminal alkynes, primary amines, ketone, vinyl, chloroalkanes, and bromoalkanes for the red dye adsorption process [67, 68]. The investigation of Khataee et al. [25] illustrated that malachite green was adsorbed by the functional groups on *Vaucheria* sp. such as methyl, terminal alkynes, C=C, and C-N.

8. Conclusion

The use of dyes is increasing day by day due to their multiple applications in different industries. The discharge of dyes into the environment not only produces an aesthetically unpleasing effect but also creates serious environmental concerns. Remediation of dye-contaminated wastewater by phycoremediation technology has gained much emphasis in recent years. Accordingly, phycoremediation technology plays a vital role principally in developing and underdeveloped countries due to the advantages such as ease of availability, high efficiency, cost effectiveness, large specific surface area, environmentally friendly, and chemical and physical stability. Algae have different functional groups on their cell wall such as amino, carboxyl, hydroxyl, and phosphate groups, which are responsible for the dye removal process. Various operational conditions such as solution pH, contact time, initial dye concentration, adsorbent dosage, and temperature are the crucial elements for the removal of dyes. Hence, these factors are taken under consideration during evaluating the performance of algal capacity regarding that of dye abatement. Despite the progressive developments in alga-based removal technologies, considerable limitations still exist which demand future research for the utilization of holistic phycoremediation removal techniques in an effective manner on a large scale.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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