

Review



# Diversity of Synthetic Dyes from Textile Industries, Discharge Impacts and Treatment Methods

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**Abstract**: Natural dyes have been used from ancient times for multiple purposes, most importantly in the field of textile dying. The increasing demand and excessive costs of natural dye extraction engendered the discovery of synthetic dyes from petrochemical compounds. Nowadays, they are dominating the textile market, with nearly  $8 \times 10^5$  tons produced per year due to their wide range of color pigments and consistent coloration. Textile industries consume huge amounts of water in the dyeing processes, making it hard to treat the enormous quantities of this hazardous wastewater. Thus, they have harmful impacts when discharged in non-treated or partially treated forms in the environment (air, soil, plants and water), causing several human diseases. In the present work we focused on synthetic dyes. We started by studying their classification which depended on the nature of the manufactured fiber (cellulose, protein and synthetic fiber dyes). Then, we mentioned the characteristics of synthetic dyes, however, we focused more on their negative impacts on the ecosystem (soil, plants, water and air) and on humans. Lastly, we discussed the applied physical, chemical and biological strategies solely or in combination for textile dye wastewater treatments. Additionally, we described the newly established nanotechnology which achieves complete discharge decontamination.

Keywords: synthetic dyes; classification; textile industries; discharge; treatment methods; nanotechnology

## 1. Introduction

The worldwide developmental process influenced all fields of life by providing rapidity, efficacy and comfort. However, it has also engendered side effects related to biosphere pollution coming from uncontrolled pollutant discharge from all sorts of industries, especially those manipulating harmful and recalcitrant compounds [1]. Particularly, the dye industries generate huge amounts of hazardous wastewater routinely [2]. Dyes are used for the coloration of several materials such as textile fibers, paper, cosmetics, tannery,



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**Copyright:** © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). leather, food, pharmaceutical products, etc., [3,4]. Before 1856, dyes were derived from natural sources only. The increasing demand and excessive costs of natural dye extraction engendered the discovery of the first synthetic dye aniline (mauveine) in 1856 by a chemist named Perkin [5]. This purple dye gave a stable and uniformly distributed color when applied to silk [6]. Dying industries depended on synthetic dyes ever since and started to expand globally, attaining nearly  $8 \times 10^5$  tons of synthetic dyes produced per year [7,8]. Notably, the textile industry accounts for ~75% of the global dyestuff market and involves around ten thousand different dyes used for printing and/or coloring multiple types of fabrics [9,10]. Textile industries are mostly located in developing countries such as India, Bangladesh, Sri Lanka and Vietnam, where they enhanced employment capacity, building the economy and foreign exchange earnings [11,12]. However, these countries do not fully respect effluent discharge norms because of their poor wastewater treatment systems. They often reject large quantities of untreated or partially treated dye effluents, eventually resulting in huge environmental pollution [13,14]. Thus, our study aims to give a comprehensive survey on textile synthetic dyes and the discharged effluents of textile industries. It focuses on introducing synthetic dyes and their classification in the first part. It delimits the impacts of these dyes on the ecosystem and human beings in the second part and discusses the available treatment methods of the released textile industry wastewater in the last part.

## 2. Classification of Dyes

Textile industries produce fibers to form yarn, which is converted to fabric [15]. They use dyes in different ways on textile fabric (Figure 1). For instance, dyeing is the process of coating the textile fiber with dyes uniformly. Printing is the application of dyes in a specific area of the fabric. Bleaching is the removal of dye color (decolorization) from textile fibers, and finishing comprises crosslinking, softening and waterproofing [7]. Dyes are classified depending on their origin into two main categories. Natural dyes, which have been known ever since ancient times, are derived mainly from plants; and synthetic dyes are artificially synthesized from chemical compounds. Synthetic dyes are divided into three groups based on the nature of the manufactured fiber. These are cellulose fiber dyes, protein fiber dyes and synthetic fibers dyes [4,16].



Figure 1. Schematic chart of synthetic dye classification.

#### 2.1. Cellulose Fiber Dyes

Cellulose fiber originates from plants such as linen, cotton, ramie, rayon, lyocell and hemp (Figure 1). These types of fabrics give perfect dyeing results with reactive dyes, direct dyes, indigo dyes and sulfur dyes [17].

#### 2.1.1. Reactive Dyes

Reactive dyes constitute the major class of cellulose fiber dyes and work well with some protein fibers (Figure 1). They are known for their high pigmentation, permanent effect, facility of manipulation under a wide temperature range and versatility due to diverse reactive groups able to form covalent bonds with multiple fibers [10,14].

#### 2.1.2. Direct Dyes

Direct dyes are very affordable, yet tend to remain in an aqueous form rather than binding to cellulose fibers (they can be used with certain synthetic fibers as well). Thereby, they are combined with inorganic electrolytes and anionic salts in the form of sodium sulfate (Na<sub>2</sub>SO<sub>4</sub>) or sodium chloride (NaCl) to enhance their fabric binding capacities (Figure 1). Thus, it is recommended to wash them in a cold cycle and with fabrics of the same color [18].

#### 2.1.3. Indigo Dyes

The indigo or dark blue color belongs to the classification of vat dyes, which are originally not soluble in water but became soluble after an alkaline reduction (Figure 1). The textile dyeing process occurs with the water-soluble or leuco form of indigo, then this form oxidizes under air exposure and returns to its original insoluble or keto form to ensure a perfect bonding of the dye to the fabric. The indigo dyes are mostly used in blue denim dyeing, which explains their production in huge amounts around the world [19,20].

#### 2.1.4. Sulfur Dyes

Sulfur dyes constitute a small, yet important class due to their excellent dyeing properties, ease of application and low cost (Figure 1). They have a complex structure with a disulfide (S–S) bridge. They belong to the vat dye classification; thus, they are reduced from the keto to the leuco form via sodium sulfide utilization. Leuco sulfur becomes soluble in water to achieve the dyeing purpose [21,22].

## 2.2. Protein Fiber Dyes

Protein fibers such as silk, cashmere, angora, mohair and wool originate from animal sources (Figure 1). They are susceptible to high pH levels; hence they are dyed using a water-soluble acid dyestuff to obtain a molecule of an insoluble dye on the fiber [23]. Acid dyes encompass azo dyes as the most important group followed by anthraquinone, triarylmethane and phtalocyanine dyes [24,25].

## 2.2.1. Azo Dyes

Azo dyes account for the largest category (60-70%) of the total synthetic dyes industry due to their versatility, cost-effectiveness, simplicity of utilization, high stability and high intensity of the color [26,27]. This dye has a prominent chromophore (-N = N-) structure, ensuring the solubility of the dyes in water and its attachment to the fiber [28,29]. Azo dyes are classified into three groups (mono, di and poly) depending on the number of azo groups in their structure (Figure 1). These groups are attached to an aromatic or heterocyclic compound on one side and an unsaturated heterocycle, carboxyl, sulphonyl, or aliphatic group on the other side [30,31].

#### 2.2.2. Anthraquinone Dyes

The class of anthraquinone is extensively used in textile dyeing industries; the red dyestuff particularly has been used for a long time [32]. These dyes are known for their

solubility in water, bright colors and excellent fastness properties (Figure 1). The antharaquinone structure could constitute junctions with azo dyes [33,34].

#### 2.2.3. Triarylmethane Dyes

The triphenylmethane dyes are widely applied in the textile industry for either dyeing wool and silk protein fibers when formed of two groups of sulfonic acid (SO<sub>3</sub>H). They can be used as indicators if they contain only one sulfonic acid (SO<sub>3</sub>H) auxochrome in their chemical structure (Figure 1). These dyestuffs are known for their solubility in water and their wide and intense color range [17,35].

## 2.2.4. Phtalocyanine Dyes

The phtalocyanine family of dyes is synthesized by a reaction between the 1,4-Dicyanobenzene compound with a metallic atom (Nickel, Cobalt, Copper, etc.) to produce green and blue shades (Figure 1). They have multiple inherent properties such as good colorfastness to light, resistance to oxidation, solubility in water and chemical stability [36,37].

## 2.3. Synthetic Fiber Dyes

Synthesized fibers are composed of spandex, polyester, acrylic, polyamide, polyoacetate, polypropylene, ingeo and acetate fabrics (Figure 1). They are used in 60% of global fiber production due to their wide application range. These fibers are dyed using direct dyes, basic dyes and disperse dyes [38,39].

## 2.3.1. Disperse Dyes

Disperse dyes are the smallest molecules among all dyes. These dyes are insoluble in water but stable under high-temperature exposure (Figure 1). The high-temperature dyeing solution is a mixture between the dyestuff powder and the dispersing agent [40–42].

#### 2.3.2. Basic Dyes

Basic dyes are also called cationic dyes because they transform into colorful cationic salts responsible for dyeing the anionic fiber textile [43]. These dyes are susceptible to light; thus, they are strictly used for dyeing paper nylon and modified polyesters. Their principal structures are cyanine, triarylmethane, anthraquinone, diarylmethane, diazahemicyanine, oxazine, hemicyanine, thiazine and hemicyanine [10] (Figure 1).

## 3. Characteristics and Impacts of Synthetic Dyes

Synthetic dyes are mostly derived from petrochemical compounds, they are commercialized in liquid, powder, pastes, or granule forms [16]. They are endowed with multiple potentialities such as fast and consistent coloration with different classes of fabrics as mentioned in the above section, a wide range of color pigments and shades, facility of manipulation, stability over several external factors and economical energy consumption [13]. Therefore, the majority of synthetic dyes cause harmful impacts when discharged in non-treated or partially treated forms in the environment [44,45]. Several reports have mentioned that up to 15% of applied dyes escape the textile fibers and are released into wastewater, and that the procedures of textile dyeing (dyeing, fixing, washing, etc.) consume a massive amount of water. Hence, a huge volume of improper discharge is rejected continuously [46,47]. Dye effluents contain high biological and chemical oxygen demand (BOD and COD) and they are very rich in organic and inorganic pollutants such as chlorinated compounds, heavy metals, sulfur, nitrates, naphtol, soaps, chromium compounds, formaldehyde, benzidine, sequestering agents and dyes and pigments [48,49]. Several toxic elements remain in the wastewater even after certain treatment processes [50,51]. Thereby, they cause multi-contamination effects on air, soil, plants and water resources, in addition to severe human diseases [52].

## 3.1. Harmful Impacts on Soil and Plants

Liquid and solid wastes discharged from textile industries contain dyes, plastic, polyester, fibers, yarns and other hazardous materials as mentioned in the above section (Figure 2). These polymeric compounds have been responsible for the pollution of local landfill habitats and agricultural fields, especially in developing countries. This soil pollution engenders plant growth inhibition by causing oxidative stress, lowering the protein content, photosynthesis and  $CO_2$  assimilating rates [7,53].



Figure 2. Direct and indirect impacts of textile dyes on several substrates.

#### 3.2. Harmful Impacts on Air

Textile dye industries release toxic gases like sulfur, formaldehyde, oxides of nitrogen, volatile compounds, particulate matter and dusts distinguished by an unpleasant smell (Figure 2). This air pollution could affect humans (workers and customers), animals, the final product and the environment [52,54,55].

#### 3.3. Harmful Impacts on Water

The main damage caused by the industrial dye effluent effects the receiving water bodies (including seas, rivers, lakes, natural ponds and streams) and spreads over large distances causing damage to other forms of life [49]. The wastewater contains multiple toxic materials and its color result from the discharge of several dyes; it is noticeable and very recalcitrant even in low concentrations (>1 mg/L), yet the average concentration of textile effluent dye reaches about 300 mg/L [56]. The dark color and high turbidity of these effluents interfere with sunlight transmission through water, decrease the amount of dissolved oxygen and disturb the pH level (Figure 2). These factors lead to several ecological impacts on the aquatic system such as the inhibition of photosynthesis in aquatic plants, low biodegradability by aerobic microorganisms and harmful effects on the food chain [57,58]. Water is highly susceptible to pollution compared to the other areas, and it is also hard to determine the pollution level in aquatic systems. The water may appear clear even though the pollution may reside for long periods in sediments and fish [16,44].

## 3.4. Harmful Impacts on Humans

Dye products and by-products existing in wastewater discharge or the dust produced inside the textile industry pose serious damages and long-lasting health impacts to human beings (Figure 2). They affect several vital organs (brain, kidney, liver, heart) and systems (respiratory, immune, reproductive) of the human organism [59,60]. Diseases may occur either directly through inhalation such as respiratory problems, asthma, allergy, nausea, or skin and eye irritation and dermatitis, or indirectly through the food chain such as tuberculosis, cancer, hemorrhage, gene mutations, and heart disease [2,61].

## 4. Applied Strategies for Textile Dye Wastewater Treatment

Textile industries use huge amounts of water in the dyeing processes, making it hard to treat the enormous quantities of wastewater discharge (Figure 3). The latter is composed of various organic and inorganic pollutants as described in the previous section [45,62]. Therefore, several countries have imposed rules to reach a standard before the effluent is released into the ecosystem or is reused for other purposes. For this to happen, physical, chemical and biological technologies have been employed to protect the environment and human health from discharge problems. These strategies could be implemented solely or in combination to obtain effective results [13,30,63].



Figure 3. Treatment strategies of textile dyes discharges.

## 4.1. Physical Treatment Process

The physical treatment (Figure 3) of industrial wastewater involves conventional processes such as adsorption, filtration, ion exchange and oxidation [16].

#### 4.1.1. Adsorption

This approach is an efficient and easy operation applied when the discharge effluent contains a large variety of dyes [56]. An appropriate adsorbent is selected depending on its affinity and ability of regeneration. Adsorbent materials are known to be pricy (e.g., activated carbon), which allowed scientific researchers to find alternative adsorbent materials

such as bentonite clay, fly ash, wheat residue, date stones, nanoparticles, etc., (Figure 3). They are affordable and effective in wastewater treatment [64,65]. This treatment strategy is achieved through the attachment of the pollutants to the specific surface area of the adsorbent material [66,67]. The achievement of dyes decolorization by adsorption with activated carbon was extensively studied. For instance, malachite green was adsorbed with curcuma-based activated carbon [65], with tetraethylenepentamine activated carbon [66] and with carbon coated layered double hydroxide [64]. Rhodamine B was successfully adsorbed by ordered mesoporous carbon and commercial activated carbon and by treated rice-husk based activated carbon Acid red 4 adsorption occurred via the activated carbon. Other adsorption methods were applied to remove the Basic fuchsin by bottom ash and deoiled soya and with mussel shell biomass waste and Acid blue 25 with shrimp shells (Table 1).

Textile Dyes	Treatment Methods	Type of the Treatment Methods	References
Malachite green	Adsorption with curcuma-based activated carbon	Physical	[65]
	Adsorption with tetraethylenepentamine functionalized activated carbon		[66]
	Penicillim ochrochloron	Biological -	[67]
	Pandoraea pulmonicola YC32		[68]
	Enterobacter asburiae XJUHX-4TM		[69]
	Flavobacterium caeni sp.		[70]
Crystal violet and malachite green	Adsorption with carbon-coated layered double hydroxide	Physical	[64]
Crystal violet	Surfactant modified magnetic nanoadsorbent	- Physical	[71]
	Adsorption with bentonite-alginate composite		[72]
	Adsorption onto TLAC/Chitosan composite		[73]
	Ozonation	Chemical -	[74]
	Electrociagulation		[75]
	Agrobacterium radiobacter	Biological -	[76]
	Diaporthe schini		[77]
Acid yellow	Adsorption on flakes of chitosan	Physical -	[78]
	Fenton oxidation		[79]
	Electrocoagulation using iron electrolites	Chemical	[80]
Rhodamine B and Acid yellow	Adsorption by ordered mesoporous carbon and commercial activated carbon	Physical	[81]

Table 1. Examples of textile dyes and their treatment methods.

Textile Dyes	Treatment Methods	Type of the Treatment Methods	References
Rhodamine B	Adsorption by micro and nano-particles of ZnO	Physical	[82]
	Adsorption with treated rice husk-based activated carbon		[83]
	Ozonation	Chemical	[84]
Basic violet and Acid blue 93	Pseudomonas putida	Biological	[85]
	Candida krusei	Biological	[86]
Basic violet 3	Fenton oxidation	Physical	[87]
Dusic violet o	Ag, ZnO and bimetallic Ag/ZnO alloy nanoparticles		[88]
Reactive Black 5 and Reactive red	Nano zerovalent iron treatment	Physical	[89]
Reactive red 195	Electro-fenton	Physical	[90]
Des stiers and 2	Adsorption sludge	Physical	[91]
Reactive fed 2	Pseudomonas sp. SUK1	Biological	[92]
Reactive red 180	Citrobacter sp. CK3	Biological	[93]
Reactive red 198	Catalytic ozonation	Chemical	[94]
Reactive red 120	Ozonation	Chemical	[95]
	Micrococcus glutamicus NCIM-2168	Biological	[96]
Reactive green	White rot fungus		[97]
	UV/H2O2 advanced oxidation process (AOP)	Physical	[98]
	Electrooxidation on Ti/IrO2-SnO2-Sb2O3	Physical	[99]
	Electrochemical oxidation		[100]
	Adsorption with calcium hydroxide		[101]
Indigo carmine	<i>Trametes hirsuta</i> laccase production	Biological	[102]
	Phanerochaete chrysosporium manganese peroxidase production		[103]
	<i>Bacillus amyloliquefaciens</i> laccase production		[93,104]
Anthraquinone, indigo and triphenylmethane	Ganoderma sp. En3	Biological	[105]
A aid rad 27	Armillaria sp. F022	Biological	[106]
Acid red 27	Chitosan adsorption	Physical	[107]
Acid red 131	Eectrochemical coagulation	Chemical	[108]
Acid red 73	Coagulation		[109]
Methyl violet, basic fuchsin and their mixture	Biosorption using fungal biomass	Biological	[110]

Table 1. Cont.

Textile Dyes	<b>Treatment Methods</b>	Type of the Treatment Methods	References
Basic fuchsin	Adsorption by graphene oxide/zinc oxide (GO/ZnO) nanocomposite	- Physical	[111]
	Adsorption by bottom ash and deoiled soya		[112]
	Adsorption on alkali-activated diatomite		[113]
	Adsorption with mussel shell biomass waste		[114]
	Electrochemical oxidation	Chemical	[115]
	Phanerochaete chrysosporium	Piological	[116]
	<i>Leptothrix</i> sp.	Diological	[117]
	Fenton oxidation		[118]
Amido black 10B	Adsorption with zeolite		[119]
Annuo black 10D	Adsorption with polyaniline/iron oxide composite	Physical	[120]
	Adsorption usingpolyaniline/SiO2 nanocomposite		[121]
Direct red 28	Electrocoagulated sludge	Chemical	[122]
Direct red	Oxidation with photo-Fenton		[123]
Direct red 23	Adsorption with PAN/PVDF composite ananofibers	Physical	[124]
Direct red 31 and Direct orange 26	Biosorption by rice husk	Physical	[125]
Direct red 89 and Reactive green 12	Biosorption	Physical	[126]
Direct Blue 1 and Direct Red 128	Biosorption using Trametes versicolor	Biological	[127]
	Acinetobacter sp. AVLB2	Biological -	[128]
4-nitroaniline	Candida sp. AVGB4		[129]
	RGO-Ni nanocomposite	Physical	[130]
Acid Blue 92	Coagulation/Flocculation	Chemical	[131]
	Ozone based oxidation	Physical	[132]
Acid Blue 113	Electrocoagulation	Chemical	[133]
	Nanofiltration	Physical -	[134]
	Adsorption by clay materials		[135]
Congo red	Ozonation	Chemical	[136]
	Aspergillus niger	Biological	[137]
Congo red	Bacillus cohnii	Biological	[138]
Remazol orange	Pseudomonas aeruginosa	Biological	[139]
Remazol brilliant orange 3R	CdO–ZnO nanofibers	Physical	[140]
Reactive blue 13	Pseudomonas sp.	Biological	[141]
Reactive orange 16	Ozone oxidation	Physical	[142]

Table 1. Cont.

Textile Dyes	<b>Treatment Methods</b>	Type of the Treatment Methods	References
Remazol Black-B	Adsorption on waste orange peel	Physical	[143]
Brilliant blue G	<i>Galactomyces geotrichum</i> and <i>Bacillus</i> sp.	Biological	[144]
Remazol brilliant blue and orange	Peel adsorption	Physical	[145]
Brilliant blue R	Adsorption with orange peel and spent tea leaves	Physical	[146]
Acid orange 7 and Remazol black 5	Biosorption	Physical	[147]
Orange 2	Adsorption by row and chemically modified brown macroalga	Physical	[148]
Acid blue 25	Biosorption with shrimp shells	Physical	[149]
Acid orange 8	Pd-Ni bimetallic nanoparticles	Physical	[150]
Basic blue 3	Pd-Ni nanoparticles supported on activated carbon	Physical	[151]
Rhodamine B	palladium-supported zirconia-based catalytic degradation	-	[152]
Acid red 4	Adsorption with activated carbon	Physical	[153]

#### Table 1. Cont.

## 4.1.2. Nanoparticle Utilization

Nanotechnology is the science of nanoscale materials (size  $\leq 100$  nm); it has attracted numerous researchers due to the great potentials of nanoparticles in removing textile dyes [68]. Nanoparticles have a wide range of classification [69], they are chemically stable, have a large surface area, can be prepared physically, chemically and biologically (microbes and plants), and are cheap and eco-friendly [70]. The shape, size, structure, purity and arrangement of nanomaterials matters in the process of decolorization [71]. Nanoparticles are mainly applied directly to wastewater to adsorb rejected dyes on their surface for further elimination [72]. They could also be used in filters, membranes and carbon nanotubes for further regeneration and reutilization in dye wastewater discharge cleaning [73]. This original methodology is taking place as a significant, large-scale solution for textile industries due to the speed of cleaning and reduction of certain pollutants to near-zero levels [74]. Nanomaterials could be synthesized based on plant extracts [75,76], metals and metal oxides [77,78], carbon [79] and nanocomposites [80]. However, it is important to know that the elimination of nanoparticles after water purification needs to be addressed [81]. Multiple scientific works covered dye degradation using nanocomposites including graphene oxide/zinc oxide (GO/ZnO) [111], polyaniline/SiO<sub>2</sub> [121] and RGO-Ni [130]. Umar et al. [150] used Pd–Ni bimetallic nanoparticles to treat Acid orange 8 (Table 1).

## 4.1.3. Filtration

Filtration is a membrane-based separation process (Figure 3) that involves popular techniques such as reverse osmosis, ultrafiltration and nanofiltration, to allow the acquisition of reusable water and recycled dyes [82–84]. The concept of these procedures consists of transporting industrial wastewater throughout several membranes differing by mesh size and separation mechanisms to finally obtain clean water [85,86]. Dye waste water treatment by the membrane filtration method was reported by [154–177], he used nanomembrane filtration. Liang et al. [178] coupled electro-fenton reaction with membrane filtration method with iron nanoparticle reduction to remove various classes of dyes [179]. This technology

proved to be prominent in dye removal, but it has some drawbacks such as permanent change to the membrane, the generation of foul odors and insoluble wastes, which implies further processing [87,88].

#### 4.1.4. Ion-Exchange

Ion-exchange adsorbents are introduced in the wastewater in either solid or liquid form, they are used to bind harmful anions or cations of the opposite charge, and in turn, release an equivalent amount of non-harmful hydrogen ions [89,90]. The review paper of Hassan and Carr [180] addressed strategies for reactive dye removal by applying the ion-exchange method [180–182]. Cationic dyes were also removed from water throughout the ion-exchange method [183,184]. Raghu and Basha (2007) combined the ion-exchange with chemical or electrochemical methods to remove dyes from textile wastewater [185]. This technique is great for the elimination of toxic and soluble pollutants (Figure 3) from effluent water; however, its use has been limited because of its high cost [91].

#### 4.1.5. Oxidation

Advanced oxidation processes (AOPs) have been extensively applied for textile dye degradation due to their powerful ability to oxidize a wide range of synthetic dyes and other complex pollutants existing in textile effluents [92,93]. They include catalytic oxidation, which is the process of active radical production (hydroxyl or sulfate radicals) on a particular catalyst surface [94,95]. In the Fenton reaction example, hydrogen peroxide is the oxidant and iron ions are the catalyst (Figure 3). This reaction occurs in an acid medium (pH  $\sim$  3) to allow the decomposition of the hydrogen peroxide into hydroxyl free radicals acting as strong oxidants [96]. Other types of Fenton techniques are also applicable such as electro-Fenton, photo-Fenton and sono-Fenton, with the possibility of a combination of them [97]. Interestingly, the acid yellow 17 [79], Basic violet 3 [87] and Amido black 10B [118] dyes were treated by the Fenton oxidation process. Otherwise, Reactive orange 16 was treated by the ozone oxidation method [142], Reactive green was removed using UV/H<sub>2</sub>O<sub>2</sub> AOP [98] and indigo carmine was subjected to the electrochemical oxidation [100]. The main drawbacks of the oxidation technique are the generation of hazardous by-products in cases of incomplete oxidation and the possibility of sludge formation [98,99].

## 4.2. Chemical Treatment Process

The most common chemical treatment processes (Figure 3) are coagulation, flocculation and ozonation [56]; they are utilized for contaminant elimination and in particular, those released in textile wastewater [100].

#### 4.2.1. Coagulation/Flocculation

Coagulation and flocculation (Figure 3) are the simplest chemical methods for the pretreatment of textile dye effluent [101,102]. This technique enables the elimination of suspended insoluble materials by adding charged chemical colloids (Aluminium sulfate (Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>), Iron (III) chloride (FeCl<sub>3</sub>), Iron (II) sulfate (FeSO<sub>4</sub>), alum, lime, etc.) provoking coagulation and settling with oppositely charged particles in the polluted water [103]. It is worth noting that Crystal violet [75], Direct red 28 [122] and Acid yellow [80] dyes were removed throughout the electrocoagulation technique. Acid red 73 was treated by coagulation [109] and Acid blue 92 was removed by the coagulation/flocculation technique [131]. The provoked residues and the necessity to use subsequent treatments to ensure the elimination of the remaining soluble contaminants are the major limits of this technique [65,104].

#### 4.2.2. Ozonation

This approach uses ozone as a strong oxidizer, its cleaning properties allow it to eliminate toxic textile effluent compounds such as azo dyes [105,106]. Other benefits of ozone, when used in the gaseous state (Figure 3), are no fluctuation of the volume of wastewater and no generation of sludge [107]. In this respect, the literature has multiple examples on the degradation of dyes including Reactive red 120 [95], Acid blue 92 [132], Rhodamine B [84] and Crystal violet [74] using the ozonation method. The main shortcomings of this process are that it is highly susceptible to many factors (pH, salts, temperature, etc.), it can release toxic compounds and its cost is elevated [103].

#### 4.3. Biological Treatment Process

The process of biodegradation/bioremediation occurs naturally via a wide variety of adapted microorganisms such as bacteria, fungi, algae and yeast existing within the wastewater and/or the polluted area [108,109]. This process could also be induced on a laboratory scale by isolating and screening appropriate microorganisms, succeeded by a scale up to allow for textile effluent treatment and decolorization [3,110,111]. The responsible microorganisms for biodegradation (Figure 3) use several techniques such as adsorption, biosorption, bioaccumulation, alleviation, elimination, or mineralization of the harmful wastewater molecules into non-harmful or even beneficial products [49,112]. This technique gained major attention for being environmentally friendly, safe, clean (no sludge), and economic, because it could be combined with other technologies [113,114]. Bacteria [115] and fungi [6] are the principal contributors to wastewater degradation due to their ability to produce degrading enzymes (oxidoreductases, hydrolases, oxygenases, ligninases, peroxidases and laccases) responsible for breaking down recalcitrant molecules [116–119]. It is noteworthy to cite the role of extracellular laccase produced by multiple fungi such as Neurospora crassa and Phanerochaete chrysosporium in the degradation of multiple dyes [186–206]. Al-Jawhari et al. [207] also mentioned the ability of few filamentous fungi in the treatment of crystal violet and methylene blue dyes [154–214]. Otherwise, Bacillus subtillis, Proteus sp. and Streptococcus sp. proved to be useful in azo dyes degradation [207,215]. Lysinibacillus sp. RGS proved to decolorize and detoxify the Remazol red dye [216–219]. Nonetheless, natural bioremediation is a time-consuming method, the induced biodegradation is not readily reproduced and elevated concentrations of toxic compounds may prevent the growth of existing or introduced microorganisms [8,102].

## 4.4. Combinatorial Treatments

All the above-mentioned treatments proved effective in alleviating textile wastewater toxicity (Figure 3). However, achieving complete decontamination is a major challenge. Thus, a multistep treatment process is needed to obtain the best possible results [50]. The example of three treatment steps was well explained by Ghaly et al. [17]. It combines several treatment methods, starting with chemical methods to allow for the elimination of solid contaminants [17,87,120]. The secondary treatment is achieved using microorganisms to reduce the COD and BOD rates, to remove turbidity and to convert the generated sludge from the primary treatment into non-harmful products [121]. In the tertiary treatment, physical methods are applied to ensure the total decontamination of the textile wastewater and its safe reuse or release in the environment [122] (Figure 3).

## 5. Conclusions

This study delimited the harmful impacts of synthetic dyes and other pollutants existing in textile effluents when discharged in non-treated or partially treated forms in the first section, and introduced physical, chemical and biological treatments and their efficiency when applied solo or combined to give rise to clean and reusable water, in another section. The future of textile dyeing industries promises wider expansion, which will see producers seeking more applications. This rapid development generates large pressures on governments, who have to be more aware of the consequences of the uncontrolled discharge of textile effluents on all sorts of life and to impose severe regulations on textile and synthetic dye industries (such as the use of environment-friendly dyes and fibers, developing employed treating techniques, and reducing water and energy consumption). Nanotechnology application in wastewater purification has a promising future due to the versatile properties of nanoparticles in reducing costs and time of wastewater cleanup operations. This strategy could be combined with any of the above-mentioned techniques to ensure the best possible results of textile dye decontamination and water reutilization for other purposes.

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