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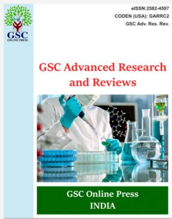

(Review Article)

Comparison of dye wastewater treatment methods: A review

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Abstract

Wastewater is produced by numerous dyes producing and dye consuming industries in their process activities especially the textile industry. These effluents become toxic and harmful to the living things and the environment if not properly treated before being discharged to the environment. In recent decades dye wastewater has been becomes a growing water pollution problem because it is one of the most difficult to treat. To put an end to this problem, viable, efficient, and sustainable method of treatment of dye wastewater and color removal needs to be established. Several research papers have been done over the years on various treatment method of dye wastewater with evolving options; this paper is to bring together both the conventional and new methods. Some of the conventional and new methods researched over the years include activated sludge, coagulation, adsorption, membrane separation processes and electrochemical process etc. Although there is currently no uniform standard or method of treatment universally adopted, many countries have put in place allowable limits of composition of dischargeable wastewater. This paper seeks to explore which methods are highly efficient, produces manageable and recyclable waste and a combination/hybrid treatment option of these methods to achieve maximum color removal.

Keywords: Adsorption; Coagulation; Color removal; Dye Wastewater; Hybrid treatment; Membrane separation

1. Introduction

Due to modernization and population growth, demand for beauty and color continues to increase. To meet this demand, many industries such as textile, leather, paper, and plastic use dyes to color their products and consume substantial volumes of water. As a result of process inefficiency from the industries, about 10-15% of dye are lost into the waste stream during production and is discharged as large volumes of dye wastewater into the environment. The release of this effluent without further treatment into water bodies results in ecosystem damage, water shortage and degeneration [1, 2, 3, 4]. The direct discharge of these effluent into natural water sources results in damages to the ecosystem and induction of mutagenic effects on mammals' organs, which greatly threatens human health [5]. Dyes are very toxic, stable, highly visible in trace amount, and not easily biodegradable. Dyes can be natural or synthetic and are classified into acid, basic, disperse, reactive, direct, vat, metal complex, sculpture and mordant dyes. There are over 10, 000 dyes used in textile production with nearly 70% being azo dyes which has a complex structure and synthetic in nature [6]. The dye wastewater has high alkalinity, biochemical oxygen demand (BOD), chemical oxygen demand (COD), total dissolved solids (TDS) and low biodegradability [7]. It is easy to identify the presence of dye in water bodies because it is aesthetically unpleasant, and the quality of water as perceived by the public is considerably influenced by the color. The highly colored component of dye obstructs the reoxygenation capacity of the water bodies and hinders sunlight penetration, thereby disrupting biological activity in aquatic life.

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Hence the aim of the paper is to provide an overview of the different types of dye, process stages in the textile industry, health and environmental impact of dye, conventional and new technologies in wastewater treatment and possible combination of the dye wastewater treatment methods.

2. The dye industry

Annually, an estimated 700, 000 tons of color is manufactured from about 100, 000 commercial dyes and globally about 3000-4000 kilotons of wastewater generated from dye producing and dye-utilizing industries. Among the various source of dye wastewater, the textile industry is largest contributor of about 54% of dye effluent polluting the environment. This account for more than half of the worldwide industrial dye effluent produced. The dyeing industry contribute up to 21% while the paper and pulp industry, tannery and paint industry, and dye manufacturing contributes up to 10%, 8% and 7% respectively to the overall through their various process activities. The distribution is represented in Table 1 [8, 9].

The unit process in the textile industry includes sizing, desizing bleaching, dyeing, and printing with some leftover dye at the process completion [10]. The leftover dye is because only about 80% of the dye and other chemical used are absorbed by the materials to be colored. The dye effluent contains many other unsafe chemicals such as hydrogen peroxide, caustic soda and many others as listed in table 2. Table 2 lists the processes, the percentage of leftover dye wastewater generated from each process, effluent composition, and wastewater characteristics.

Table 1 Industrial sources of dye wastewater [8]

Industry	Percentage contribution
Textile	54%
Dyeing	21%
Paper and pulp	10%
Tannery and paint	8%
Dye producers	7%

Table 2 Percentage and composition of dye wastewater from textile manufacturing [8, 11]

Unit process	% of dye waste water discharge	Wastewater composition	Wastewater characteristics
Desizing	21%	Starch, carboxymethyl cellulose, polyvinyl alcohol, waxes, ammonia	High BOD and COD, dissolved solids
Scouring	52%	Waxes, caustic soda, surfactants, soda ash	High COD, dissolved solids
Bleaching	62%	Caustic soda, hypochlorite, surfactants, acids, chlorine, hydrogen peroxide	Alkalinity, suspended solids (SS)
Mercerizing	4%	Sodium peroxide, cotton wax	High pH, low BOD, high dissolved solids (DS)
Dyeing	85%	Reducing agents, oxidizing agents, detergents, dyestuffs, urea, wetting agents	Highly colored, Heavy metals, high BOD, low SS
Printing	13%	Gum, starch, binders, oil, reducing agents	Highly colored, high BOD, oily appearance, SS
Finishing	58%	chlorinated compounds, waxes, inorganic salts, softener	Low BOD, slightly alkaline, high toxicity

3. Dyes

Dye is natural or synthetic organic compound that connect itself to surfaces or fabrics to provide bright and lasting color. Dyes have affinity towards the substrate. Most dyes are soluble in water and generally applied to aqueous solutions although may require a mordant to improve the fastness of the dye on the fiber. During the early years, dye was obtained from natural sources which include the plants trees, lichens, and insect. Attempts to extract more dyes from brightly colored plants and flowers failed because a lot of natural dyes are unstable occurring as components of complex mixtures. Based on the inadequate availability of natural dye and the high demand for dye, the synthetic dye was developed. Synthetic dyes are derived from complex organic or inorganic compound. In general, they are produced in the dye industries from chemicals. Most dyes used today are in this category. The first synthetic dye was accidentally discovered by William Henry Perkin while looking for a cure for malaria disease in 1856. Due to low production costs and easy application to the fabric, synthetic dyes are produced in large scale, but their toxic nature is a cause for concern. Dye wastewater generated from using the synthetic dye which is usually in large volume pose serious threat to lining things and the environment at large. Natural dyes have certain advantage over synthetic dye such as no toxicity, easy extraction and purification, renewable resources, little to no effluent generation. A better option to explore would have been to revert to using natural dyes but natural dyes have its disadvantage such as requiring the use of mordant to ensure proper bonding to fabrics, poor shade productivity and poor color fastness [12]. Mordants are toxic binding agents which help natural dyes to attach to fabric and pose the same amount of risk to the environment. Synthetic dyes have complex molecules which are stable because they possess auxochrome and chromospheres which enables their water-soluble bonding and coloring characteristics.

3.1. Classification of dyes

Table 3 Classification and application of yes [8, 13, 14]

Dye type	Water solubility	Chromophoric groups	Application	Examples
Acid	Soluble	Azo, anthraquinone, azine, nitroso, triphenylmethane, xanthene, nitro.	Food, silk, leather, wool, nylon, paper, Cosmetics, printing ink, acrylics, polyamide fibers	Acid Yellow 36, Acid Orange 19
Basic	Soluble	Acridine, azo, oxazine anthraquinone, azine, cyanine, thiazine, diazahemicyanine, xanthene, triarylmethane	Silk, wool, Inks, wood, medicine, paper, straw tannin-mordant cotton, polyesters, leather	Crystal violet, Methylene Blue
Azo	A type of direct dye	Stilbene, pyrazoles, coumarin, anaphthalimides	Rayon, cotton, plastics, paints, acetate, cellulosic materials, detergents	Acid red 88, Acid orange 19
Disperse	Insoluble	Azo, anthraquinone, nitro, styryl, benzodifuranone groups	Polyester, nylon, plastic, cellulose acetate, acrylic fibers	Disperse Blue 27, Disperse Yellow 3
Direct	Soluble	Polyazo compounds, stilbenes, oxazines, phthalocyanines,	Cotton and rayon, paper, leather, nylon, wool, silk cellulose, fibers, linen,	Direct Orange 39, Direct Blue 15
Fluorescent brighteners	Mostly soluble but some insoluble	Naphthylamides, stilbene coumarin, pyrazolos	All fibers, oils, paints, plastics, soaps, detergents	4, 4′-bis (ethoxycarbonyl vinyl) stilbene
Mordant	Soluble	Anthraquinone and azo	Anodized aluminum, wool, leather,	Mordant Blue 3
Oxidation bases	-na-	Aniline black and indeterminate structures	Cotton, fur, and hair	Direct Blue

Reactive	Soluble	Azo, anthraquinone, triarylmethane, formazan phthalocyanine, oxazine	Cotton, wool, yarn, silk cellulosic, painting, polychromatic printing	Reactive Blue 5
Vat	Insoluble but soluble with alkali	Anthraquinone (including polycyclic quinones), indigoids, and carbazole	Cotton, cellulosic fibres, polyester-cotton, rayon, and wool	Vat Orange 1, indigo (vat blue 1)
Solvent	insoluble (solvent soluble), nonpolar or little polar	Azo, anthraquinone, phthalocyanine, triarylmethane	Plastics, gasoline, lubricants, oils, waxes	Solvent red 24, Solvent yellow 124
Sulfur	Insoluble	Nitro and amino groups	Cotton, rayon, polyamide fibers, silk, leather, paper,	Sulphur black 1, indophenol

Dyes are classified based on their unique chemistry, structure, way of bonding, source of materials, chemical composition, and industrial performance as shown in Table 2. They could be natural or synthetic based on the source of material used to produce them. Examples of natural dyes are Turmeric (Curcuma longa), Onion (Allium cepa), and Indigo (Indigo era tinctoria). Examples of the synthetic dyes used by the industries includes azo, disperse, Acid, basic, direct, mordant, reactive, sculpture and vat dyes with azo dyes being the most produced and utilized class of dye up to a rate of about 70% of the total worldwide usage of dye.

4. Effect of dye on health and environment

Although dye impacts color on material bringing about beautification and enhancing the products of various industry, the production and utilization processes of dye comes along with several health and environmental hazards. Long term exposure of workers to dyestuffs and other chemicals used in production and utilization of dye can lead to health hazards while chemicals must be handled with care. These chemicals include formaldehyde-based resins, ammonia, acetic acid, shrink-resist chemicals, optical whiteners, soda ash, caustic soda, and bleach. Commonly for reactive dyes, Inhalation of dye particles lead to respiratory problems (respiratory sensitization), and often it affects a person's immune system [15]. Symptoms and side effects includes itching, watery eyes, sneezing, coughing, wheezing, skin irritation and sore eyes [6]. During the dying process for treatment of cloth with boiling liquor, acid and alkalis used can lead to risk to the burns and scalds in workers are exposure occurs. Other hazards can result from chips flying from metals like chromium when it strikes workers. Aromatic amines used in dying industries can cause DNA (deoxyribonucleic acid) mutation. When using sculpture dye which requires to reducing agents like formaldehyde, exposure to reducing agents can lead to cancer on nose, lung, and brain [16]. Textile industries produce large amounts of liquid effluents and sludge which contain organic and inorganic compounds. If this effluent is not properly treated, it will prevent sunlight from penetrating through water surface to provide required oxygen by aquatic creatures [17]. This produce a visible layer above water which is aesthetically unpleasant and produce a foul odor thereby polluting the air. The dye effluent also destroys soil productivity if it finds its way to the soil.

4.1. Environmental standard for wastewater discharge

In recent years, there has been increasing environmental awareness and stricter governmental regulations about the discharge of toxic colored wastewater into the water bodies. Also, the scarcity and increasing cost of water for industrial processes has made treatment and reusing of color effluent a good alternative for industries. Based on these and other factors, dye producing and utilizing industries have to ensure wastewater discharge from there operations meet the standard quality and guideline provided by environmental regulating bodies such as the USEPA (U.S. Environmental Protection Agency) and others as designated by different countries. For textile wastewater, the main concern for the regulatory bodies are metal ions, dyes, and its colors because of their hazardous impact on human and the environment. This standards and allowable limit are presented in Table 4.

To meet the below standard and limit and mitigate cost of treatment and buying of fresh water, industries need to develop efficient method for contaminant and color removal as well as consider the option of reusing the wastewater produced.

Table 4 Standard for Dye Effluent Discharge by Countries [8, 16, 18]

Country	Temperature (°C)	pH	BOD (mg/l)	COD(g/l)	SS (mg/l)
United State	42	6. 5-8. 5	30-45 mg/l		30-45
Nigeria	40	6. 0-9. 0	30–50	60–90	25
Uganda	35	6. 0-8. 0	50	100	10
Malaysia	40	5. 5-9. 0	50	100	100
Thailand	40	5. 5-9. 0	20–60	20–60	120–400
Global Limit	Below 42	6. 0 – 9. 0	Below 30	Below 50	Below 20

5. Dye wastewater treatment

There are several methods that can be used for color removal from industrial dye effluent. But because there are various dyes available and industrial effluent contains other chemicals aside the dye, many treatment methods may not efficient singly and may need to be combined with other treatment method to achieve maximum color removal. Dye wastewater treatment method can be categorized into three namely physical, biological, and chemical treatments [19, 20, 21, 22].

5.1. Physical method of dye removal

Conventional physical method of dye removal includes coagulation-flocculation, adsorption, ion exchange, reverse osmosis, membrane filtration and nano filtration or ultra-filtration. The physical method of treatments is the most used because they and easy to set up and operate and are mostly efficient for color removal.

5.2. Biological method of dye removal

Another cheap and easy to operate alternative for dye removal is the biological method so it is largely used in most countries for dye wastewater treatment. Biological method involves the bacterial degradation of dye by bacteria, fungi, yeast, and algae during aerobic and anaerobic process [23]. However, this method is ineffective because it is unable to removal dye and toxic although it is economically feasible, environmental-friendly, generates less volume of sludge and has the capacity to treat the chemical oxygen demand (COD) in wastewater [19, 24]. Other biological method includes adsorption by microbial biomass, algae degradation, enzyme degradation, fungal cultures, and microbial cultures.

5.3. Chemical method of dye removal

This method involves the application of chemistry theories to achieve dye removal. In comparison to the biological and physical methods, this is not a preferred method by industries because more expensive to setup and operate. It involves high energy consumption and high investments in chemicals and reagents. Part of its disadvantage is the issue of disposal of secondary toxic pollutant produced during the operation of dye removal [19]. This method includes advanced oxidation process, electrochemical destruction, Fenton reaction dye removal, oxidation, ozonation, photochemical and ultraviolet irradiation. Coagulation and flocculation treatment processes have been used to remove color and organic pollutants from dye wastewaters [25, 26]. Low cost adsorbents have been used to remove color from various dye containing wastewaters [27, 28, 29, 30]. One type of low cost adsorbents is agriculturally based adsorbents, which have been employed for decolorization of dye containing combined wastewaters [31, 32].

Table 5 Merits and demerits of dye wastewater treatment methods [8, 11, 12].

Methods	Merits	Demerits
Physical Treatment		
Adsorption by activated carbon	Excellent ability to remove a wide variety of dyes. Adsorbent regeneration	Costly and expensive
Ion exchange	Regeneration prevents loss absorbent. High quality water output	Effective for few numbers of dyes

Electrocoagulation	Inexpensive and feasible	Production of large sludge
Membrane Filtration	Effective for all dye type, water recovery and reuse	expensive to setup, generate concentrated sludge
Irradiation	Effective and optimized at laboratory scale	Requires lots of oxygen,
Reverse osmosis	Production of clean water, useful for water recycling and effective for wide variety of dye	Needs high amount of pressure and expensive
Biological Treatment		
Enzyme degradation.	It is nontoxic, reusable, and inexpensive and highly efficient	Unreliable enzyme production
Adsorption by microbial biomass	Some dyes have high affinity that allows them to bind with microbial biomass	Effective for limited number dyes
Aerobic-anaerobic (conventional) method	Cheap and effective for decolorizing a wide variety of dye	Produces sludge, Yields methane and hydrogen sulphide as by-products
Microbial cultures (mixed bacterial)	Takes between 24-30 hours to decolorize	Not effective for all dyes
Chemical Treatment		
Fenton reaction	Removes toxic, good for removal of both soluble and insoluble dyes	Iron sludge generation, long reaction time and ineffective for disperse and vat dyes.
Photochemical	No sludge and foul production and effective for dye removal	Generation of several by-products
Oxidation	Simple application with short reaction time	Expensive and requires catalyst for efficient removal. Difficulty activating H_2O_2
Electrochemical destruction	No sludge build-up or chemical consumption	Produces hazardous materials, and high cost of electricity

Table 6 Performance of Various Dye Wastewater Treatment Methods

Treatment Method	Dye	Conditions and Results	(%) Removal	Reference
Physical				
Adsorption				
Adsorption by activated carbonrice processing waste, peanut shell, Aspergillus niger and laccase	Aniline blue	Maximum dye removal by activated carbon at adsorbent dose of 1 g/L, temperature of 60oCat10min contact time	98	[33]
Adsorption by Fe-based metal-organic frameworks (Fe-MOFs	Rhodamine B, Congo	23. 3855 mg/L of Congo red, 22. 7365 mg/L of Orange II and 17. 9973 mg/L of Rhodamine B in 200 mL solution within 300 min of	99. 57 95. 9899. 38	[34]

	Red, Orange II	treatment with natural light at 15oC		
Average removal percentage 98. 23				
Ion exchange				
Anion-exchange with sulfonic acid and phosphate groups	Methyl violet 2B	Synthetic dye wastewater made up of 0. 03 g/L methyl violet 2B and 2 g/L Na2SO4 at pH 3 and 100 °C	93	[35]
Anion-exchange by Lewatit MonoPlus MP 500 resin	Acid Orange 7	The experiment had maximum adsorption capacity at 1004. 4 mg/g, 0. 5 g anion dosage, contact time is 3 h, dye concentration of 10 mg/L, pH of 5 and temperature of 45 °C.	87	[36]
Average removal percentage 90				
Coagulation/flocculation				
Coagulation/flocculation using ferric chloride sludge from water treatment plant	Acid red 119	Surface methodology (RSM) was used to optimize initial pH (3. 5), coagulant dosage(236. 68mg) and initial dye concentration (65. 91) for dye	96. 53	[37]
Coagulation/flocculation using polyaluminum chloride and polydiallyldimethyl ammonium chloride	Multiple dyes wastewater	Dye removal at the optimal dosage of PAC/PDDA=400/200 ppm and pH>3	90	[34]
Average removal percentage 93. 27				
Irradiation				
Irradiation by TiO2/H3PW12O40 film Excited under Solar-Like Radiation	Alizarin red	Maximum dye removal was achieved when contact time is 240 min, initial dye concentration of 25 mg/L	89. 8	[38]
Irradiation by periodate ion concentration	Basic Red 46, basic yellow 28	Highest TOC removal efficiency obtained at pH 3. 0 using 5 mM periodate ion in the presence of 1 g/L TiO2 for both dye solutions in 3 hours illumination	76	[39]
Average removal percentage 82. 65				
Reverse Osmosis				
Reverse osmosis	Acid red, reactive black reactive blue	Dye concentration at 65 mg/L, temperature at 39°C and pressure at 8 bars	97. 2 99. 58 99. 9	[40]
50 Dalton of reverse osmosis	Reactive, disperse direct, and acidic dyes	Ideal parameters include a temperature of 35°C contact time of 2 h, a dye concentration of 50 mg/L, a pressure of 7. 5 bars, flowrate of 10 L/min and dye concentration of 100 mg/L.	99. 6 98 95	[41]
Average removal percentage 98. 21				

Biological				
Enzymatic degradation				
Enzymatic degradation using soybean peroxidase and Luffa acutangula peroxidase	Azo dye methyl orange	Maximum dye decolorization in 1 h incubation at 30 °C using 2 mM of hydrogen peroxide, 0. 5 mL crude soybean peroxidase and 30 mg L−1 dye at pH 5. 0 and in 40 min at 40 °C using 2 mM hydrogen peroxide, 1. 5 mL crude luffa peroxidase and 10 mg L−1 dye at pH 3. 0.	81. 4 75. 3	[42]
Enzymatic degradation by white rot fungus Datronia sp. KAPI0039	Reactive blue 19, reactive black 5	Decolorization of 1000mg/l reactive Blue 19at 2% (w/v) Datronia sp. at pH 5 and 600mg/l reactive Blue	86 88. 01	[43]
Average removal percentage 82. 68				
Adsorption by microbial biomass				
Enterobacter dissolvens AGYP1 and Pseudomonas aeruginosa AGYP2	Acid Maroon V	Maximum dye removal(absorbance) after 6 hours contact time at dye concentration of 100mg/l	96%	[1]
Low-cost biosorbent (P. animale)	Textile dye	Maximum dye removal (absorbance) at 93. 16 mg/L, 45 °C, 1440 minutes contact time and 4 g/L for pH	99. 66	[44]
Average removal percentage 97. 83				
Aerobic-anaerobic (conventional) method				
Sequential anaerobic–aerobic treatment	Reactive Red 195	system operated at θH=18 h, Temperature of 19–22 °C, 3000 mg l−1 initial COD concentration and 100 mg l−1 dyestuff concentration to obtain over 85% decolorization efficiency in anaerobic reactor, 15% color removal and 90% COD removal in aerobicunit	90	[45]
Decolorizing anaerobic/aerobic sequencing batch reactors	Acid Red 88	The sequential anoxic–aerobic treatment of synthetic dye wastewater (SDW) feed having 100 mg L−1 of AR-88 dye resulted in the 98% color and 95% COD removal.	98	[46]
Average removal percentage 89				
Chemical				
Electrochemical Oxidation				
Pulse electrochemical oxidation for treating recalcitrant dye wastewater	Indigo Carmine, Alizarin Red S, and	Treated with PbO2/Ti anode and Box-Behnken designs. This can save energy consumptions up to 35. 5%, 40. 1%, and 47. 9% for IC, ARS, and MO, respectively	88. 4	[47]

	Methyl Orange			
Electrochemical oxidation using Ti/Ru0. 3Ti0. 7O2 composite anode	Acid brown 98	Synthetic wastewater containing 0. 1 M NaCl treated at pH 3 and 20 mA cm-2. 67% TOC removal with 0. 1 M, NaCl as an electrolyte, 20mAcm-2 current density, under 60 minutes of electrolysis	90	[48]
Average removal percentage 89. 2				
Fenton reaction				
Fenton reaction through Fe (II)/H2O2 reagents	Cibacron Red FN-R	20 mg l−1 Fe (II)reagent and 250 mg l−1 H2O2, applied for irradiation time of 90 min in a 24-h-cycle	80	[49]
Fenton's oxidation	Direct Blue 71	Optimal conditions for the decolorization and COD removal of DB71 at pH = 3. 0, Fe2+ = 3 mg L−1, H2O2 = 125 mg L−1, temperature 20 -60 °C and 20 min reaction time	94 50. 7 COD	[50]
Average removal percentage 87				

6. Hybrid method of dye removal

The textile wastewater contains dye and many other contaminants which are sometimes nonbiodegradable in nature. Exploration of a hybrid system considering the advantage of each process as listed in Table 5 may help achieve maximum efficiency of dye wastewater treatment. Systems that can be combined to give maximum efficient needs to be investigated depending on the nature of the wastewater sample. For example, anaerobic and filtration systems were combined for domestic wastewater treatment. To reduce the space requirement, retention time, investment, operation, and maintenance costs a simple filtration step as a post treatment solution for anaerobic processes is employed. The removal efficiencies of TSS, COD and FC (faecal coliform) for combined system were 93%, 87% and 93%, respectively, against TSS (45%), COD (38%) and FC (78%) removal by UASB (upflow anaerobic sludge blanket) reactor alone.

7. Conclusion

Although there is no uniform standard globally, many countries have put in place strict limit for wastewater discharge and industries can explore both the conventional and new technologies to meet this standard. Moreover, a lot of research has been done on dye wastewater treatment processes individually but more research into the hybridization can help to increase efficiency of treatment. A combination of the adsorption, coagulation-flocculation and the filtration process will efficiency remove color, COD from dye wastewaters.

Compliance with ethical standards

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Disclosure of conflict of interest

The authors, Bukola M. Adesanmi, Yung-Tse Hung, Howard H. Paul and Christopher R. Huhnke, declare no conflict of interest.

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