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Review

Batik Effluent Treatment and Decolorization—A Review

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Abstract: Batik is a piece of woven cloth decorated with beautiful patterns and designs and has become a signature product of the Malay Archipelago, including Malaysia and Indonesia. Batik industry consumes a large volume of water and produces a large amount of wastewater during the boiling process and dyeing process, both for hand-drawn (batik lukis) and block-printed (batik cap) batik. The release of colored effluents that contain a large number of dyes and chemicals can harm the environment and become a human health concern, particularly in south east Asian countries. Therefore, treatments of batik effluent are very crucial and have caught a lot of attention from researchers. The color removal is a major challenge, especially from this industry, as up until now there is no single and cost-effective treatment that can effectively decolorize as well as treat the dye effluent. Since batik is part of the textile industry, most treatment methods have been adapted from textile effluent treatment. Here, we review a variety of textile wastewater treatment techniques to make a good consideration of selecting the most appropriate method to be applied in batik wastewater. First, we briefly review the batik process, including the potential dyes that are mostly used in batik processing. Secondly, we describe all possible techniques and their performance to reduce dye concentration and decolorization. Finally, we review all advantages and disadvantages of these techniques for domestic and industrial applications.

Keywords: batik; dyes; physical treatment; chemical treatment; biological treatment

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1. Introduction

The word batik originally comes from a combination of the Javanese word “amba” and “nitik”, which mean “writing” and “drop”, respectively [1]. Batik has been produced since the 17th century and is considered to be a national heritage of the Malay world for both Malaysia and Indonesia. In 2009, UNESCO recognized batik as a Masterpiece of Oral and Intangible Heritage of Humanity. The industry of batik started in small-scale workshops, often in backyards or alongside the river. This unique craft has since become commercialized on a massive scale, which gives a positive contribution to the economic growth due to the high level of demand for batik in both local and international markets [2]. The utilization of the product is not strictly only on clothing, but has also been extended to handbags, shoes, book marks, and pillow covers [3].

The process of batik begins by applying melted wax onto plain white cloth to create patterns and designs (known in the local dialect as canting), dyeing the cloth, and finally by undertaking multiple washing procedures [4]. Contamination from batik printing can occur in four different stages of the production process, as shown in Figure 1. However, the most polluted wastewater is from the final stage, i.e., after the final wash, as it contains both wax and resin.

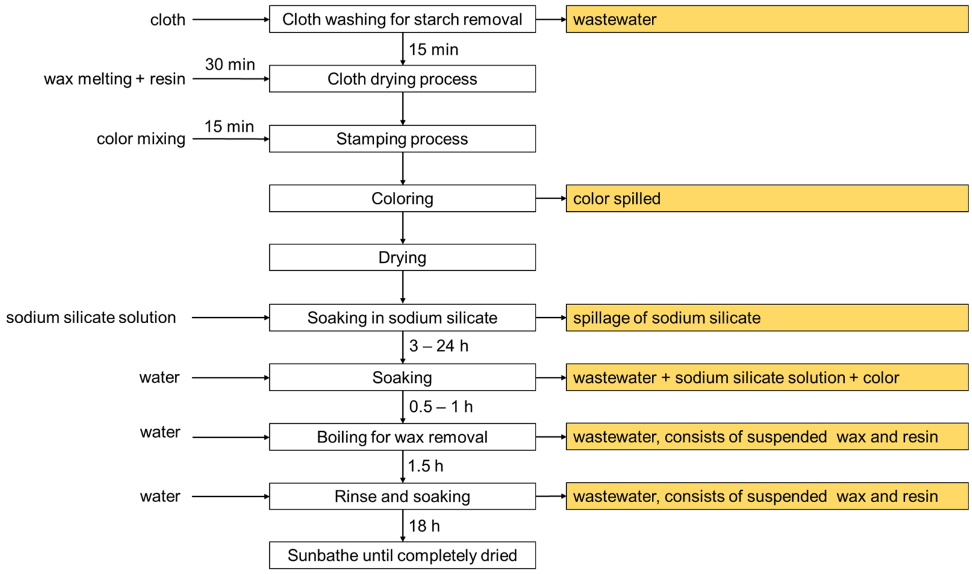


Figure 1. Flowchart of the batik process, where highlighted boxes show the possible sources for water pollution from batik making. This flowchart was modified from [5].

These processes generate large amounts of effluents with a high concentration of pollutants due to the chemicals used mainly during the dyeing process. These effluents, particularly when flowing untreated, will contribute to water pollution and degrade the quality of the environment. The small-scale batik workshops usually have no or limited treatment processes, whereby the wastewater is directly discharged into the water streams. As such, the scale of contamination is as high as in other industrial discharges, whereby continuous contamination and no implemented effective actions would lead to the deterioration of the water stream [6].

As the treatment of batik effluent is very crucial, the manufacturers have to be responsible to treat this effluent by applying some treatment technologies to ensure that the effluent is safe to be discharged. Typically, to ensure human health and also the environment, the effluent discharge should be secured and maintained by the standards set by the authorities. Therefore, the Malaysian Department of Environment has made guidelines and standards for details on industrial effluent discharge. This review paper aims to give comprehensive information on batik wastewater treatment that comply with these regulations, starting with the general characteristics of effluent from batik factories, the dangers posed to the environment by releasing untreated batik effluent, and ending with a list of potential technologies for treating this wastewater.

2. Production of Batik

Batik is a piece of woven cloth decorated with various patterns and designs. Batik lukis (hand-drawn batik) and batik cap (block-printed batik) are the two most commonly used processes in the art of batik making. Batik lukis undergoes the waxing process completely by hand using a canting tool, while batik cap is waxed using a copper stamp. The process begins with the preparation of fabrics, raw materials, such as waxes and dyes, as well as suitable tools, such as canting and block stamps [7].

The exclusivity of one batik piece depends on whether it was made through batik lukis or stamping. The high-end batik requires a skilled designer and is usually determined by its smooth melted wax drawing (as shown in Figure 2) and the fine technique applied during coloring. The colored fabric is then boiled to remove all waxes and finally, the batik is dried under the mild sun in the drying process.

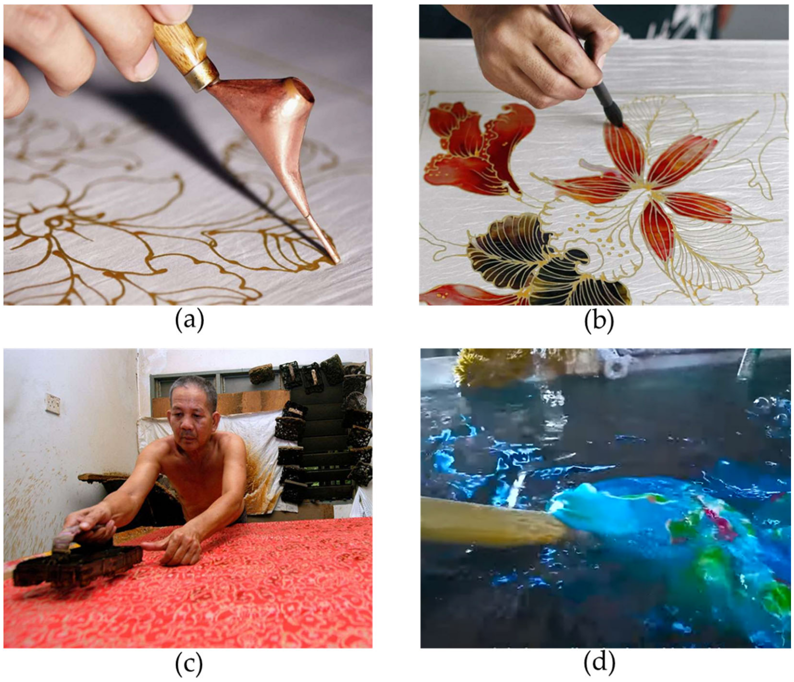


Figure 2. Schematic activities of Batik production: (**a**) canting process; (**b**) paint dyeing process; (**c**) block printing process; (**d**) wax removal process [7].

Due to the repetitive process of waxing and dyeing to create a more elaborate and colorful design, the increment usage of synthetic or chemical dyes has resulted in the high concentration of chemicals and suspended solids in the wastewater, particularly during washing and boiling processes [8]. In addition, the boiling process also consumes a large amount of water to remove the wax, which puts stress on the water requirement. The effluents from the boiling process are normally discharged without proper treatment or with minimal treatment and are one of the point sources of high organic pollutant wastewater [9].

The types of dyes and chemicals used in the textile industry are found to differ depending on the fabrics used. The raw materials for textiles are natural and synthetic fibers. Batik is commonly made from natural fibers of cellulose (i.e., cotton) and protein (i.e., silk). Cellulose fibers are obtained from plant sources, such as cotton, rayon, and linen, whilst the protein fibers are made from animals and include wool, angora, and silk [10].

In the early days, batik was authentically made from naturally resourced dyes, such as plants and insects. However, due to the large-scale demand of batik, both synthetic and/or chemical dyes are more commonly used compared to natural colors. Common dyes used on cellulose fibers include reactive dyes (e.g., Remazol, Procion MX, and Cibacron F), direct dyes (e.g., Congo red, direct yellow 50, and direct brown 116), naphthol dyes (e.g., fast yellow GC, fast scarlet R, and fast blue B) and indigo dyes (e.g., indigo white, Tyrian purple and indigo carmine) [10,11]. Reactive dyes are the most commonly used dye to pigment cellulose fiber, such as cotton. Cotton could also be dyed using direct dyes, i.e., indigo dyes and naphthol dyes. Tables 1 and 2 show the structure of the common dyes used in batik printing.

Protein fibers are dyed using acid dyes (i.e., azo dyes, triarylmethane dyes, and anthraquinone dyes) and Lanaset dyes (of Blue 5G and Bordeaux B) [12]. As a low pH is needed to denature the proteins, acid dyes are most commonly used to dye protein fibers. The chemical reactions between the acid dye and the fibers are found to form an insoluble dye molecule on the fiber. Acid dyes are found to contain azo groups (-N=N-) in the center. Lanaset dyes are a popular group of dyes that are classified under both reactive and acid dyes. Reactive dyes are also commonly used to dye protein fibers. Silk could be dyed using acid dyes, reactive dyes, or naphthol dyes [11]. However, based on varying dye types available in the market, the most popular choice for the batik producers is the reactive dye of Remazol and vinyl sulfone [13].

Table 1. Structure of some direct dyes [14].

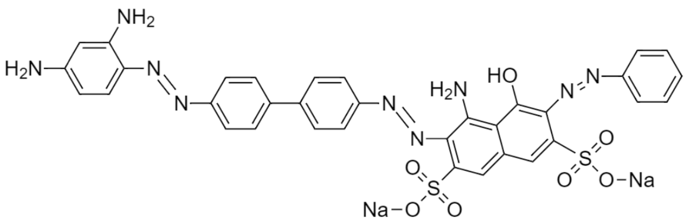
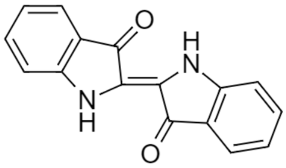
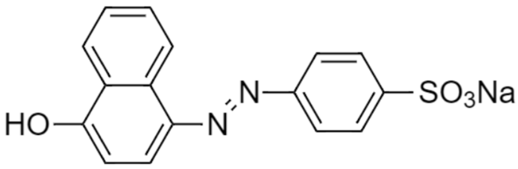
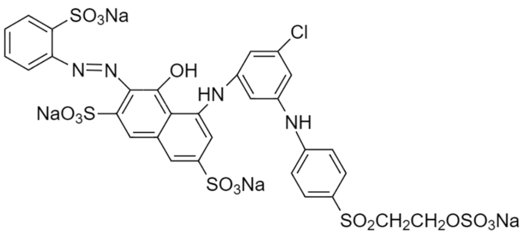
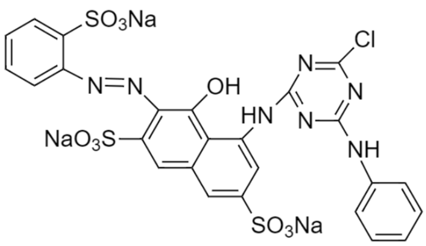
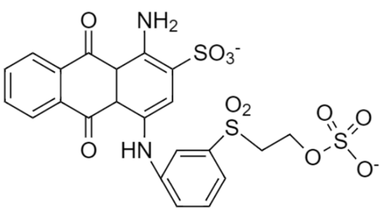
Dyes	Chemical Structure	Molecular Weight (g/mol)	Maximum Absorption Wavelength (nm)
Direct dyes (Direct Black 38)		760.8	520
Indigo dyes (Vat blue 1)		262.26	610
Naphthol dyes (Acid Orange 20)		350.3	488

Table 2. Structure of several reactive dyes [14].

Dyes	Chemical Structure	Molecular Weight (g/mol)	Maximum Absorption Wavelength (nm)
Vinyl sulfone (Reactive Red 239)		1050.37	555
Procion MX (Reactive Red 3)		774.05	532
Remazol (Reactive Blue 19)		626.5	592

3. Characteristics of Batik Effluent

The characteristics of batik effluents vary and depend on the types of textiles and the chemicals used. Batik wastewater effluents contain high pollutants causing damage to the environment and human health, including suspended and dissolved solids, biochemical oxygen demand (BOD), chemical oxygen demand (COD), chemicals, odor, and color [15]. The wastewater discharge may also contain a significant level of grease, wax, surfactant, and silicate [16]. Not only that, but the probability of the presence of trace metals, such as Cr, Co, Ni, As, Cu, Pb, and Cd is highly likely because they come from dyes used in production, especially azo dyes that are very commonly used in batik production [17]. Table 3 gives an idea of the general characteristics of raw batik wastewater effluents.

Table 3. General characteristics of untreated batik wastewater effluents compared to the common characteristics of textile wastewater, and the permitted value set by the Malaysian Department of Environment.

Parameter	Batik Effluent [1]	Textile Effluent [2]	Permitted Value [3]
pH	7.65–12.5	5.5–10.5	5.5–9.0
Dissolved Oxygen (DO) (mg/L)	1.7–2.25	-	3
BOD_5 (mg/L)	340	80–6000	50
COD (mg/L) [4]	1600–4090	150–30,000	250
TSS (mg/L)	305	15–8000	100
Turbidity (NTU)	217	-	-

Notes: [1] from reference [18–20]. [2] from reference [21]. [3] Permitted effluent discharge is obtained for Standard B of the Malaysian Environmental Quality (Industrial Effluents) Regulations 2009 (see Table 4). [4] The value for COD discharge is as per the accepted condition for the textile industry.

To compare the level of danger posed by batik wastewater to the environment, the common characteristics are compared with the untreated textile wastewater effluents. Batik wastewater has similar characteristics to textile wastewater although the BOD, COD, and TSS values are lower. However, batik wastewater still exceeds the acceptable value set by the Malaysian Department of Environment (DOE) for industrial effluents.

The degree of pollution from batik wastewater depends on the quantity of water and chemicals used in the dyeing process. The more stable complex molecular structure of the dyes enables them to remain in the water for an extended period [22]. Dyed effluents reduce the depth of penetration of sunlight into the water, which in turn decreases photosynthetic activity and results in a detrimental concentration of dissolved oxygen (DO) [23]. Because aquatic organisms need light to develop, any deficit in the light reaching the aquatic life results in an imbalance in the ecosystem. Dyes also increase the BOD of the receiving water and in turn reduce the re-oxygenation process, and hence affect the growth of photoautotrophic organisms [24].

Dyes in solution have a direct effect on humans by causing diseases, such as hemorrhage, ulceration of the skin, nausea, severe irritation of the skin, and dermatitis [25]. Not only that, but continuous exposure to textile dyes in powder form has resulted in lung and skin irritations, headaches, and nausea [11]. Commonly used dyes, such as disperse blue, disperse violet, disperse orange, and disperse red, were found to have the presence of mutagenic agents and were toxic to humans, animals, plants, and microorganisms [26,27].

A high concentration of suspended solids from the batik wastewater creates bonding with oily scum, and interferes with the mechanism of oxygen transfer in the air-water interface. Inorganic substances in the textile effluents make the water unsuitable for use due to the presence of an excess concentration of soluble salts. These substances, even in a lower quantity, are found to be toxic to aquatic life.

Heavy metals in batik wastewater can harm the ecosystem, even in low concentration. Swarnkumar et al. [28] discovered that heavy metals contained in textile wastewater, such as cadmium, chromium, lead, arsenic, and zinc, affected aquatic plant growth and the development of chlorophyll content in seedling reduction. Sampling of freshwater contaminated with textile wastewater in Nigeria revealed that the dyes in wastewater had contaminated the freshwater with mercury, cadmium, and arsenic. The researchers also observed contamination of arsenic, lead, mercury, and cadmium in the sediments of the freshwater. This heavy metal pollution resulted in the extinction of thousands of archaea, bacteria, and fungi species in this ecosystem [29].

Table 4. Acceptable conditions for industrial effluents or mixed effluent discharge set by the Malaysian Ministry of Natural Resources and the Environment [30].

No.	Parameter	Unit	Standard	
			A	B
1.	Temperature	°C	40	40
2.	pH Value	-	6.0–9.0	5.5–9.0
3.	BOD at 20 °C	mg/L	20	50
4.	Suspended Solids	mg/L	50	100
5.	Mercury	mg/L	0.005	0.05
6.	Cadmium	mg/L	0.01	0.02
7.	Chromium, Hexavalent	mg/L	0.05	0.05
8.	Chromium, Trivalent	mg/L	0.20	1.0
9.	Arsenic	mg/L	0.05	0.10
10.	Cyanide	mg/L	0.05	0.10
11.	Lead	mg/L	0.10	0.5
12.	Copper	mg/L	0.20	1.0
13.	Manganese	mg/L	0.20	1.0
14.	Nickel	mg/L	0.20	1.0
15.	Tin	mg/L	0.20	1.0
16.	Zinc	mg/L	2.0	2.0
17.	Boron	mg/L	1.0	4.0
18.	Iron (Fe)	mg/L	1.0	5.0
19.	Silver	mg/L	0.1	1.0
20.	Aluminum	mg/L	10	15
21.	Selenium	mg/L	0.02	0.5
22.	Barium	mg/L	1.0	2.0
23.	Fluoride	mg/L	2.0	5.0
24.	Formaldehyde	mg/L	1.0	2.0
25.	Phenol	mg/L	0.001	1.0
26.	Free Chlorine	mg/L	1.0	2.0
27.	Sulfide	mg/L	0.50	0.50
28.	Oil and Grease	mg/L	1.0	10
29.	Ammoniacal Nitrogen	mg/L	10	20
30.	Color	ADMI *	100	200

Notes: * ADMI-American Dye Manufacturers Institute. Standard A is for discharge in catchment areas, whereas standard B is for discharge in any other inland waters.

4. Discharge Standards for Industrial Effluents

In Malaysia, to maintain a safe and healthy environment, Environmental Quality (Industrial Effluents) Regulations 2009 Fifth Schedule (Table 4) were enacted as guidelines detailing industrial effluent discharge, including batik industry effluents. According to these regulations, industrial effluents mean any waste in liquid form or wastewater generated from many manufacturing processes, including the treatment of water supply or any activity occurring at industrial premises. The industrial wastewater to be discharged must not exceed Standard A or Standard B, where Standard A poses a stringent requirement and is applied where discharge points are in catchment areas upstream. Table 5 shows the acceptable values of COD in the wastewater released by specific industries, including batik workshops, as a textile industry.

Table 5. Acceptable chemical oxygen demand (COD) for the discharge of industrial effluents for specific trade or industry sector regulated by the Malaysian Ministry of Natural Resources and the Environment [30].

Trade/Industry			Unit	Standard A	Standard B
(a)	Pulp and Paper Industry				
	(i)	Pulp mill	mg/L	80	350
	(ii)	Paper mill (recycled)	mg/L	80	250
	(iii)	Pulp and paper mill	mg/L	80	300
(b)	**Textile Industry**		**mg/L**	**80**	**250**
(c)	Fermentation and Distillery Industry		mg/L	400	400
(d)	Other Industries		mg/L	80	200

Notes: Based on Seventh Schedule of Environmental Quality (Industrial Effluents) Regulations 2009. Standard A is for discharge in catchment areas, whereas standard B is for discharge in any other inland waters. Bold in "Textile Industry" row is to highlight that batik industry effluent is regulated as textile industry.

A few important parameters that are typically involved in batik effluent monitoring consist of pH, color, BOD, COD, ammoniacal nitrogen, nitrate nitrogen, and suspended solids. The effluent sample will be tested through in-situ and laboratory analyses to determine the results for each parameter. Because the point of discharge of batik effluents must comply with the regulations, the batik entrepreneurs must take responsible steps by applying cleaner production practices, particularly by implementing wastewater treatment technology in their respective factories.

5. Wastewater Treatment Techniques

Batik wastewater contains high concentrations of pollutants, such as a high intensity of color, carcinogenic dyes, and toxic heavy metals. The effluents need to be treated before being discharged so that they are safe, clean, and contain no adverse effects to the environment and human health. Wastewater treatment is the purification process of contaminated water through physical, mechanical, chemical, and biological processes. This process aims to make wastewater become more acceptable for the aquatic environment [31].

The separation of dyes from batik industry wastewater has become the focus of current environmental problems [5]. Due to this serious form of water pollution, many studies have been conducted to find an effective and economical way for the treatment of dye-containing wastewater in order to preserve the environment, as well as to meet the stringent regulations. There are a number of treatment techniques or methods that can be used for color removal in batik effluents, namely physical, chemical, and biological processing.

Physical processing includes adsorption using activated carbon and membrane filtration. Chemical processing consists of methods, such as coagulation-flocculation, Fenton reaction, and ozonation. Biological treatment is generally carried out by using a bacterial consortium to degrade dye compounds. Activated sludge is a conventional treatment process that is used to remove color and improve water quality.

5.1. Membrane Filtration

Membrane filtration is a new separation method that uses the micropores and the selectivity permeability of the membrane to filter and separate certain substances in wastewater. These materials will be separated by size using pressure and a specific pore size in the membrane. When batik wastewater is pumped through membrane modules, larger molecules, such as wax and dye that are bigger than the pore size will be retained on one side, while pure water can pass through the membrane, as illustrated in Figure 3. This technology can be used for almost any kind of separation and has a high separation efficiency, low energy consumption, easy operation with no pollution, and so on [32].

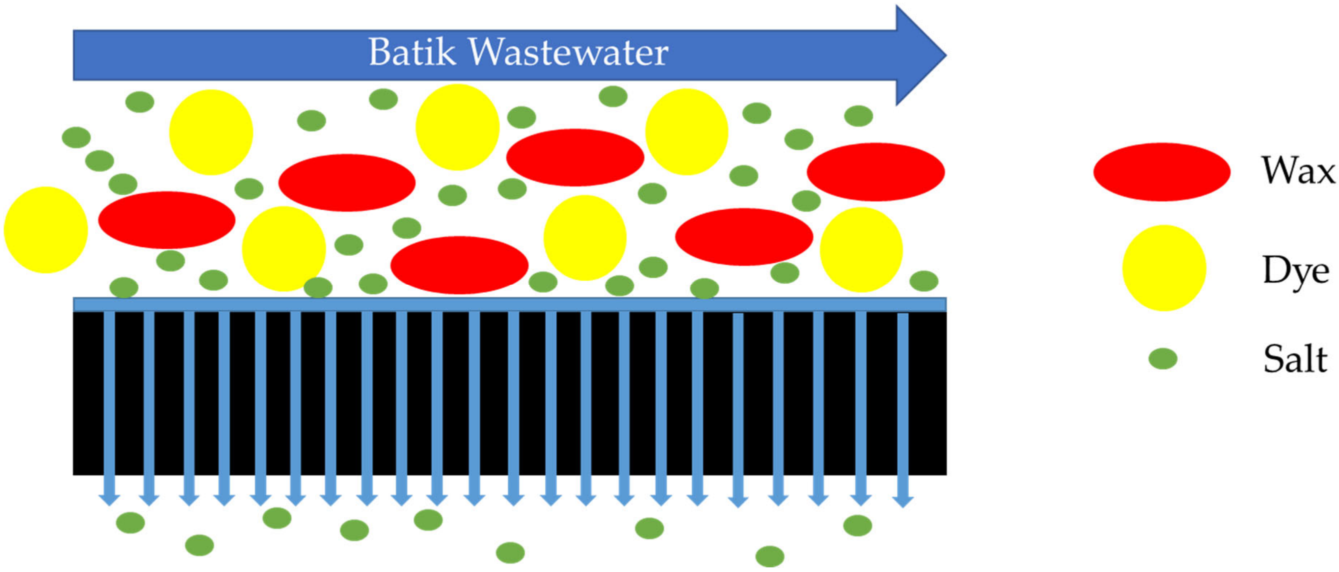


Figure 3. Illustration of contaminants separation from batik wastewater using membrane filtration (modified from [33]).

This process has the ability to clarify, concentrate, and most importantly, separate dye continuously from the effluents [34] and it also can remove color, BOD, and COD in wastewater simultaneously [35]. However, it is currently not applicable for large scale treatment, as it has some limitations, such as a short membrane lifetime, insufficient separation selectivity for some dyes, membrane fouling, high operating cost, inability for dissolved solids removal, and production of concentrated sludge [36,37]. Despite those drawbacks, it can separate many components from the liquid stream, as many categories of membranes exist. Membranes are usually categorized based on the molecular weight cut off (MWCO) and the pressure that acts as the driving force, namely reverse osmosis (RO), nanofiltration (NF), ultrafiltration (UF), and microfiltration (MF).

5.1.1. Reverse Osmosis (RO)

This technique is controlled by the diffusion process through a dense membrane with a very fine pore size. Water will dissolve in the hydrophilic membrane material under high pressure, and then diffuse through the membrane polymer structure [38]. This membrane has a retention time of more than 90% for most types of ionic compounds and produces high quality permeate. This process can perform decolorization and the elimination of chemical auxiliaries simultaneously, and also allows for the removal of all mineral salts, hydrolyzed reactive dyes, and chemical auxiliaries [37]. A laboratory experiment observed COD and color removal by RO at 99.6% and nearly 100%, respectively, for batch operation, and 91.9% COD removal with 96.4% color removal for the continuous mode [39].

5.1.2. Nanofiltration (NF)

Nanofiltration technology has been applied for the treatment of dye-containing wastewater from the textile industry. NF membranes' pores have a size of less than 1 nm with a molecular weight cut-off (MWCO) of 300–500 Da [40]. For batik dye effluents, the treatment can be carried out with the combination of adsorption and nanofiltration techniques by which the adsorption will precede nanofiltration [32]. This is because the nanofiltration will act as a screener to ensure the effectiveness of the adsorption process [41]. Nanofiltration membranes retain the low molecular weight of organic compounds, divalent ions, large monovalent ions, hydrolyzed reactive dyes, and dyeing auxiliaries [37]. Moreover, in NF, a higher rejection of dyes and other low-molecular-weight organic compounds (200 to 1000 MW) are achievable [41].

Moreover, according to [42], this process is currently not applicable for the treatment of highly concentrated and complex effluents because the contaminants trigger fouling on the membrane's surface. Furthermore, the disadvantages include the accumulation of dissolved solids and dyes, and these effluents cannot be simply discharged into a water stream without appropriate treatment [42]. Still, it is a fairly satisfactory alternative that makes this technique become favorable, in terms of compliance with environmental law [37]. Nanofiltration was proven to be able to remove more than 99% of color, suspended solids, and oil and fat from batik wastewater. Organic pollutants were also separated, resulting in COD and BOD_5 rejection rates of 87.43% and 86.23%, respectively [33]. A pilot-scale plant of two NF membranes constructed in series was able to treat batik wastewater to recover more than 80% of water to be reused [43].

5.1.3. Ultrafiltration (UF)

Ultrafiltration membrane has pores with a size of about 1 nm to 0.05 µm. This technique uses a differential pressure interval of separation membranes as the driving force, which enables the elimination of macromolecules and particles. Recent advancements in ultrafiltration have led to a better performance of removing dyes from wastewater, with a removal rate of 79.8–100% [44,45]. From research into batik wastewater conducted by [46], the optimum reduction percentage of COD can be achieved when the membranes used for separation have a pore size of 0.01 to 0.015 µm, a feed solution pH of 2, and an operating pressure of 1 atm.

The UF system alone could not treat textile effluents for water recycling, however the combination with NF could produce 14–45% of the wastewater that could be reused again without any effect on fabric dyeing parameters and quality [47]. Another alternative is for producing brackish water from a series of UF membranes to be reused in the fabric coloring process [48,49]. Ultrafiltration can only be used as a pre-treatment for microfiltration or reverse osmosis, or in combination with a biological reactor [34,50]. In the UF system, MWCO (molecular weight cut off) is important. However, the main problem affecting its widely practical usefulness is that it often requires replacement of the membrane due to irreversible fouling, especially when in high salt concentrations [51,52].

Some pre-treatment steps can be used to eliminate fouling. The presence of a high concentration of dissolved solids on the membrane surface will increase the fouling potential and decrease the membrane flux. Pre-treatment is necessary in order to separate the dissolved solids contained in the effluents and other compounds that can damage the membrane. This can be overcome by the process of clarification, lime softening, coagulation flocculation, and pre filtration [53].

5.1.4. Microfiltration (MF)

Microfiltration has pores size of about 0.1 to 5 µm and requires pressure of less than 5 bar [54]; which is suitable for treating pigment dyes in wastewater. The chemicals that are not filtered by microfiltration will remain in the wastewater at a higher feed concentration. Cake formation on the membrane surface was found to contribute to membrane fouling [55]. MF could not remove most of the color from wastewater since many soluble dyes have a smaller size than MF's pore size, although it was observed to be able to reduce nearly all turbidity [56]. Several modifications can be carried out to enhance the MF's ability for color removal, such as the addition of polyethyleneimine and graphene oxide [57], the incorporation of nano zero-valent iron (nZVI) particles, or modification with sulfuric acid, chitosan, and graphene oxide [58]. Microfiltration can also be used as a pre-treatment for nanofiltration or reverse osmosis to reduce the load [59].

5.2. Coagulation-Flocculation Treatment

This combination treatment is one of the most used methods, especially in the conventional treatment process because of its effectiveness to eliminate the insoluble dyes [60] and partly remove total suspended solids (TSSs), BOD, and COD [61]. The principle of the coagulation process is the addition of a coagulant into the wastewater followed by a general mixing of the coagulant and the pollutants to eliminate the surface electrical charges of the colloids [62]. Normally, the colloids in textile wastewater bring negative charges, so the coagulants are usually inorganic and organic cationic coagulants that bring positive charges to the wastewater (Figure 4). Examples of coagulants that are common for this treatment are aluminum sulfate, ferric sulfate, and ferric chloride [62], although natural coagulants have recently been studied for potential application [63,64]. Li et al. [65] and Adinew [66] agreed that the coagulants can also help the particle aggregation into flocks, and subsequently increase the sedimentation. The sedimentation is then removed by settling, filtration, and other physical techniques to generate sludge that is normally further treated by the dewatering process [67]. According to research by [63], coagulation removed dissolved and suspended solids in textile wastewater with a COD and color removal efficiency of up to 83.05% and 82.2%, respectively. Another study by [68] utilized bittern (a residual product from the salt crystallization process) as coagulant because of its magnesium, chloride, and sulfate ions. These ions could coagulate the heavy metals and turbidity of batik wastewater, resulting in Pb^{2+} removal of 99.3% and turbidity removal of up to 97% [69].

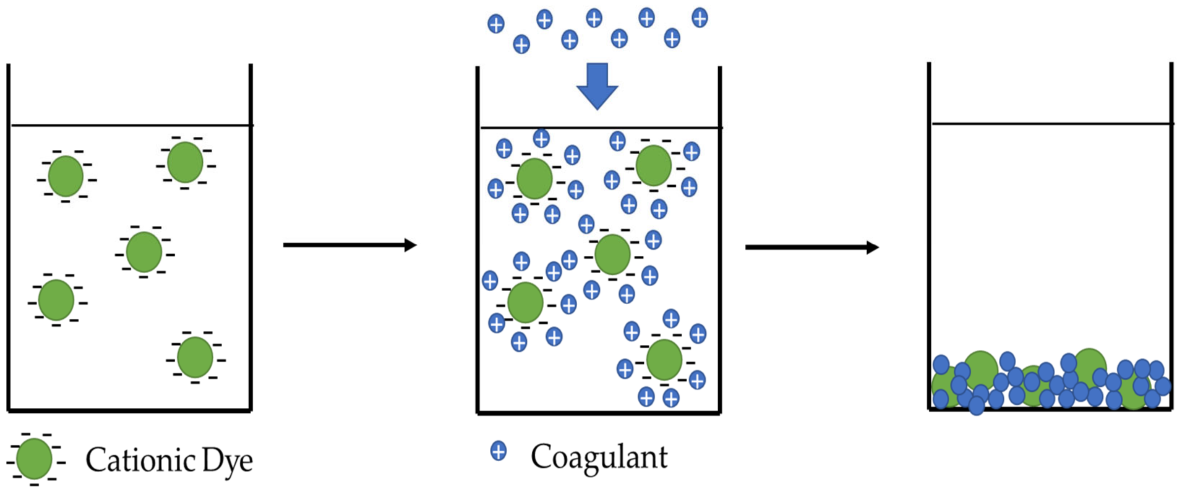


Figure 4. Illustration of coagulation of wastewater dyes by coagulants.

However, the dissolved matter in batik wastewater is not well removed by coagulation. For better removal, biological or other physical chemical processes can be used. In addition, the high chemical load in the effluent may cause the increase of sludge production and lead to incomplete dye removal. The disadvantages of this process are the high cost of treating the sludge, and there are some restrictions concerning the disposal of sludge [66]. Further to those drawbacks, Ashtekar et al. [70] mentioned that nowadays, organic coagulant polymers have been developed for color removal and they also offer advantages over inorganic coagulant polymers, such as a lower sludge production, they require less dosage, and have an improved ability for color removal.

5.3. Adsorption

Adsorption is the most used method in physicochemical wastewater treatment. It is the process by which ions and molecules that present in one phase tend to accumulate on the surface of another phase [66]. Most adsorbents are a highly porous material. In the adsorption process, basically, the wastewater is mixed directly with the porous material powder or granules and are left to pass through a filter bed containing the porous materials. This process will let pollutants bound into the surface of the solid surface [71]. This technique gives significant results as it can be used to remove different types of dye materials [72,73], including heavy metals in textile wastewater [74]. Adsorption techniques have become popular in wastewater treatment, as they are efficient in the removal of non-biodegradable pollutants to produce a high quality of recycled water [66].

Adinew [66] and Husien et al. [75] agreed that the most commonly used adsorbent is activated carbon, as so far it is still the best adsorbent to treat wastewater containing different classes of dyes. Activated carbon was reported to be able to remove COD, TSS, and color of textile effluents by 78.8%, 76.2%, and 84%, respectively, after a 20 min contact time [76]. Unfortunately, activated carbon has a low performance on adsorbing suspended solids and insoluble dyes in spite of its great activity against water-soluble dyes in wastewater. Furthermore, the wastewater might need pre-treatment before passing through the adsorption process to prevent the suspended solids from clogging the bed [77]. Activated carbon is also generally used in lower concentrations of dye wastewater treatment, or only for advanced treatment options because of its high cost, although many new low-cost adsorbents have recently been discovered [66,71,78]. One example of these low-cost adsorbents is coal bottom ash, a waste produced from coal combustion with high silica composition that could adsorb up to 41.6% TSS, 65.3% BOD, and 75.6% turbidity of batik wastewater [79].

5.4. Fenton Reaction

Wastewater treatment using Fenton reaction is carried out chemically, utilizing strong oxidizing agents and iron ions as catalysts for decomposing the oxidizing agents into radical species [80]. The strongest oxidizing agent that is commonly used is hydrogen peroxide (H_2O_2), as it can decolorize a wide range of dyes [32]. The method is based on the formation of a reactive hydroxyl radical from H_2O_2. By adding H_2O_2 to an acidic solution of pH 2 to 3 containing Fe^{2+} ions, hydroxyl radicals are formed. The hydroxyl radicals produced are very reactive and can attack organic contaminants in wastewater producing new intermediates with a lower molecular weight or even lead to the complete mineralization into carbon dioxide and water [81].

This reaction is mainly used as a pre-treatment for wastewater that contains non-biodegradable or toxic compounds, so that the biological treatment afterwards will be able to efficiently degrade the pollutants from the waste streams [80]. Higher temperatures for a Fenton reaction is desirable to boost the reaction kinetic, although contradictory results are still observed when a high temperature is applied [82]. Still, the reaction can be carried out at ambient temperature using a large excess of iron catalysts. In such conditions, the great amount of total COD removal is mainly because of the iron (III) hydroxide ($Fe(OH)_3$) co-precipitation [32].

Kusumawati et al. [46] used a hybrid process by combining Fenton oxidation and separation techniques using an ultrafiltration membrane. These researchers used this method for the treatment of batik wastewater since this combination is commonly used for wastewater with high COD and TDS content. Furthermore, this oxidation process has some influences on wastewater treatment, as it can enhance the biodegradability through the process of moving the non-biodegradable compound and dyes. The pre-treatment of batik wastewater that was conducted by [46] using Fenton oxidation reaction before the adsorption process using activated carbon, resulted in 60% to 75% of COD reduction. Although the total COD removal was quite large, the batik wastewater needed to undergo the UF system to further treat the wastewater for the better removal of pollutants. Sharma et al. [83] reported that the Fenton process had capabilities of removing various colors in simulated textile wastewater, with an efficiency between 73.86 and 81.35%. This process is limited, since textile wastewaters have a high pH and the Fenton reaction requires a low pH, resulting in the significant cost of acid addition to reach the required pH [32,66].

Moreover, at a higher pH, there is high sludge production due to the precipitation of ferric ion salts, and the process loses its effectiveness [66].

5.5. Ozonation

Ozonation or ozone oxidation is a very effective and fast decolorization treatment of water-soluble dyes, which can break the double bonds present in most of the dyes [32,66]. According to [84], it can destroy the properties of residual surfactants and can also oxidize a significant part of COD. Moreover, Ulucan-Altuntas et al. [85] mentioned that it can improve the biodegradability of effluents containing a high fraction of non-biodegradable and toxic components, through the conversion of the pollutants into more easily biodegradable compounds if these molecules need to be treated with a biological method afterwards. This process also requires pre-treatments, such as filtration, to reduce the suspended solid contents as well as to improve the efficiency of decolorization [86].

Bilińska et al. [87] used a combination of ozonation and electrocoagulation for the treatment of textile wastewater. It was shown that the ozonation effectively reduced color concentration by 95% in 60 min, and when it was coupled with electrocoagulation, the same performance could be achieved in a shorter amount of time (18 min). Toxicity of batik wastewater was reduced using ozonation in an experiment inside a 2 L batch reactor [88]. One of the biggest limitations of industrial applications of ozonation are the toxic compounds produced as byproducts of the microcontaminant mineralization process [89]. Coupled with the biodegradation process before ozonation, it has been proven to reduce the toxicity of the effluents in wastewater treatment [90]. The low mass transfer from gas (ozone) to liquid also limits the application of this technology [91]. The cost of ozonation is driven by the ozone generation technology, although it is relatively cheaper compared to the photo-Fenton process [92].

5.6. Biological Treatment

Based on [93], biological treatment systems are commonly applied to the treatment of industrial effluents because many microorganisms, such as bacteria, yeasts, algae [94], and fungi [95] are able to accumulate and degrade different pollutants, as all biological systems require a continuous input of effluents. Enzymes obtained from microbes and plants are another option of biological treatment for treating batik wastewater, since they are more resistant to rapid changes in operational conditions compared to the microbial process [96]. Biological methods are generally cheap and simple to apply and are currently used to remove organics and color from dyeing and textile wastewater [66].

Biological treatments are different depending on the presence or absence of oxygen. This can be biologically achieved with the help of microorganisms under aerobic or anaerobic conditions. Aerobic bacteria use organic matter as a source of energy and nutrients. They oxidize dissolved organic matter to CO_2 and water, and degrade nitrogenous organic matter into ammonia. Anaerobic bacteria degrade organic compounds in the absence of oxygen to produce methane and more easily biodegradable compounds, such as starch and polyvinyl alcohol. In practice, textile wastewater is usually treated through a combination of aerobic and anaerobic methods, where the anaerobic process is used to decrease the COD content rapidly, followed by the aerobic method to further polish the COD content to a very small amount [97].

5.6.1. Activated Sludge System

Activated sludge processes are commonly used for treating textile wastewater. It involves the regular aeration of the effluent inside a tank allowing the aerobic bacteria to degrade the soluble and suspended organic matter. A part of the organic matter is oxidized into CO_2 and the rest are synthesized into new microbial cells [98]. The effluents and the sludge generated from this process are separated using sedimentation; some of the sludge is returned to the tank as a source of microbes (Figure 5). A pilot-scale study on activated sludge demonstrated that it was capable of biodegrading 95% of dyes after hydraulic retention times of 5 days, and in a large scale study, the same operational parameters allowed for a dye biodegradation efficiency of 89% [99].

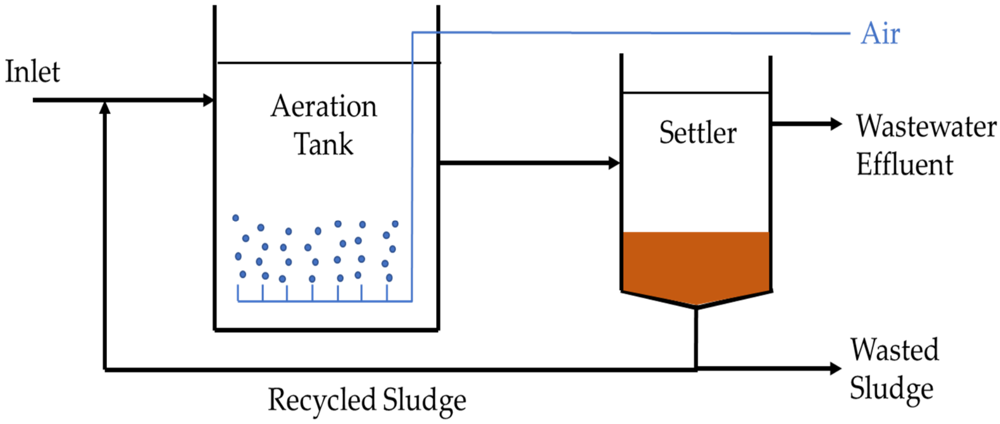


Figure 5. Schematic of the activated sludge system.

Because of the low biodegradability and toxicity of most of the dyes and chemicals used in the textile industry, their biological treatment by the activated sludge process does not always achieve great success [100]. It is remarkable that most of these dyes resist aerobic biological treatment, so adsorbents, such as kaolin, talc, or sawdust are added to the biological treatment systems in order to eliminate non-biodegradable or microorganism-toxic organic substances produced by the textile industry [101].

5.6.2. Trickling Filters

Trickling filters are another common method of secondary treatment that mostly operate under aerobic conditions. The effluents for the primary treatment are trickled or sprayed over the filter, as displayed in Figure 6. The filter usually consists of a rectangular or circular bed of coal, gravel, poly vinyl chloride (PVC), broken stones, or synthetic resins. A gelatinous film, made up of microorganisms, is formed on the surface of the filter medium. These organisms help in the oxidation of organic matter in the effluents to carbon dioxide and water [102].

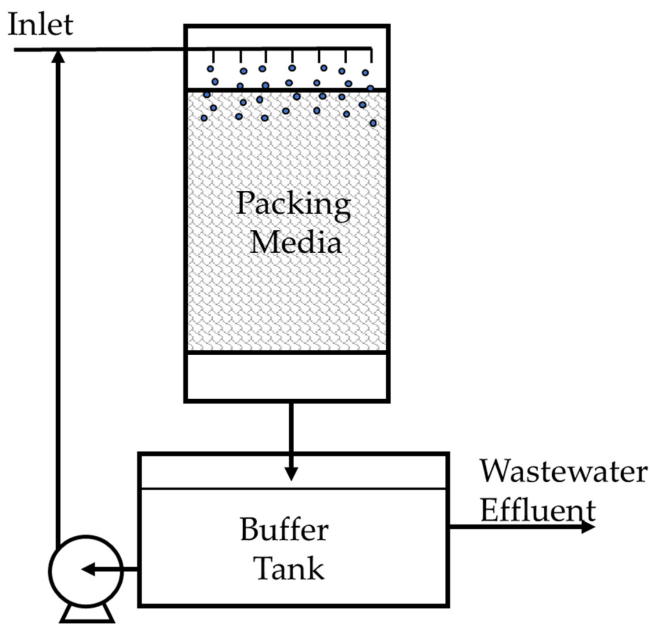


Figure 6. Schematic of the trickling filter system.

The advantages of using a bio-trickling filter over the conventional activated sludge process include a lower operational cost, less area requirement, and a well stabilized sludge, as there is no sludge bulking or floating problem. A laboratory scale trickling filter with consortia of *Pseudomonas aeruginosa*, *Alcaligenes* sp., and *Brevilbacillus parabrevis* was tested to treat simulated textile wastewater with great azo dye removal capabilities. This technology could decolorize 96.20% of reactive blue 221, 99.88% of reactive yellow 145, and 99.88% of reactive red after around 10 days. This research also found a reduction of COD (58.5–75.1%), phosphates (63.6–73.0%), and sulfates (18.9–36.5%) in textile wastewater feed [103]. Another type of bio-trickling filter using a sponge as a biomass-retaining carrier had a good performance in removing organic content, total nitrogen compounds, and color with maximum efficiencies of 93%, 56%, and 72%, respectively. The sponge facilitated the aerobic and anaerobic degradation of the dyes. It was found that *Acinetobacter* that grew in the sponge were the predominant bacteria that degrade the azo dyes [104].

5.6.3. Upflow Anaerobic Sludge Blanket (UASB)

UASB is one of the most adopted anaerobic biological technologies used in various industries due to its high efficiency, low capital expenditure, and its versatility and reliability. This technology is suitable for batik and textile wastewater because it can resist toxic compounds in the wastewater [105]. In principle, UASB operates anaerobically in a reactor where wastewater flows upwards through a "sludge blanket" containing microflora that are attached to the granular sludge bed. The configuration ensures rapid decomposition from the effective mixing between the biomass sludge and wastewater due to the generated biogas [106].

Using UASB, COD in textile wastewater could be reduced by 57.5% from 2000 mg/L, while dye concentration was reduced by 71.0%. When multiple dyes were used in the textile production, there was a decreasing performance observed due to the toxic intermediates produced from the chromogenic decomposition of the dyes, i.e., COD removal efficiency was reduced to 47.8% while color removal efficiency was reduced to 38.3% [107]. The combination of coagulation and UASB was demonstrated to increase the efficiency of COD and color removal from 70% and 81% for UASB only, to 95% and 100%, respectively [108].

5.6.4. Enzymatic Treatment

Enzymatic treatment can be carried out through a homogenous reaction by dissolving the enzymes directly into batik wastewater, or through heterogenous reaction where enzymes are immobilized into insoluble supports to facilitate enzyme recovery and reusability [96]. Examples of insoluble supports for enzyme immobilization are alginates [109], light expanded clay aggregates [110], and nanomaterials, such as carbon nanotubes, ZnO, MnO_2, and Fe_3O_4 nanoparticles [111]. Common enzymes for degrading dyes in textile wastewaters are laccases, azoreductases, peroxidases, and polyphenol oxidases [112,113]. Enzyme treatment has a high specificity and is cost effective, although enzyme recovery may pose a challenge. Immobilization can overcome this drawback but may add a diffusion barrier for the contaminants to reach the enzymes. Large scale demonstrations of enzymatic treatments are still rare, so this may add many uncertainties for its application in batik industries [111].

Laccase that was immobilized into a light expanded clay aggregate was proven to successfully degrade indigosol dyes in batik wastewater by 98.2%. The performance can be maintained even after the immobilized enzymes are used in four cycles [110]. Another study tested a crude horseradish peroxidase enzyme extracted from horseradish roots to treat textile wastewater containing Acid Orange 7 dye. A dye removal rate of 73% was obtained by this crude enzyme in a homogenous batch reactor. The removal rate was increased to 89% when the enzyme was purified by ammonium precipitation. Hydrogen peroxide could inhibit this enzyme's activity, while addition of polyethylene glycol could boost the dye removal rate [114]. When the peroxidase enzyme was immobilized in Fe_3O_4 magnetic nanoparticles, dye removal activity could be maintained in a wide range of pHs and temperature variations, even after 100 cycles. The magnetic property offers an easier separation of the nanoparticles, while the nano size could enhance enzyme activity [115].

5.7. Electrical-Assisted Treatment

Electrochemical-assisted chemical processes, such as plasma/electrical discharge, have been intensively studied in the way of finding some potential applications for environmental purposes. Currently in our daily activities, plasma has been placed in many air distributors, such as air conditioning, to destroy the presence of gaseous toxic emissions. As it has the advantage of producing very highly dense discharges, the plasma process has been found to be more effective for organic compounds, such as chloroform [116,117], carbon tetrachloride [118,119], and dichlorobenzene [120] compared to other methods, e.g., electrochemical and combustion.

Attempts to decompose organic dyes by plasma methods have emerged during the last decade and have resulted in promising results. Manoj Kumar Reddy et al. [121] researched the decomposition of methylene blue dye by using dielectric barrier discharge (DBD). When the initial dye concentration in wastewater was 100 ppm, they found that the reduction in TOC was only 20%, with methylene blue degradation reaching 94.57%. Dojčinović et al. [122] worked with four azo dyes: Reactive Black 5, Reactive Blue 52, Reactive Yellow 125, and Reactive Green 15. Those four dyes' concentrations were significantly reduced by 95% compared to the initial concentration, by the addition of peroxide with a specific energy supply of 18–25 kJ/g. Dojčinović et al. [122] reported a successful degradation of azo dye Acid Orange 7 (AO7) and antraquinone dye Acid Green 27 (AG27) under ozonation. Using the similar dye of AO7, [123] tried to optimize the utilization of pulse discharge plasma (PDP) and found that decolorization could be increased to 19.4% compared to the base-case adsorption of activated carbon. A more comprehensive decomposition experiment with a greater number of dyes by plasma discharge was conducted by [124]. Fourteen different dyes were tested and the results showed that the DBD method can be successfully used in a variety of synthetic dye polluted water treatments at low concentrations (<50 mg/L). The degradation byproducts are easily biodegradable and less toxic, and can be further oxidized in biological treatment facilities. This technique is currently shown to be very promising, but the development faces a tremendous challenge as this technique relies on electrical power generation.

6. Summary of the Advantages and Disadvantages of Physical Chemical and Biological Methods

From the review in Section 5, general advantages and disadvantages of the main treatment methods of dye removal from industrial effluents are shown in Table 6.

Table 6. Advantages and disadvantages of main physical, chemical, and biological methods of dye removal in textile wastewater (summarized from [31,111]).

Treatment Methods	Method Description	Dye Removal Performance	Advantages	Disadvantages	References
Fenton reagent	Decompose contaminants using strong oxidizing agents and iron ions as catalyst	81.35–97.8%	• Effective decolorization of both soluble and insoluble dyes; • Effective for textile wastewater with solid contents.	• Sludge generation; • Relatively long reaction time; • Effective only in a low pH.	[83,125]

Table 6. *Cont.*

Treatment Methods	Method Description	Dye Removal Performance	Advantages	Disadvantages	References
Ozonation	Dissolving ozone in wastewater that acts as a strong oxidizing agent	95–100%	• Applied in a gaseous state; • Effectively degrades pollutants in a short amount of time; • Does not produce sludge.	• Short half-life (20 min); • High cost to generate ozone; • Sometimes forms toxic and carcinogenic byproducts.	[87,126]
Membrane filtration	Utilizes pore sizes and pressure to separate contaminants in wastewater	• Reverse Osmosis: 96.4–100%; • Nanofiltration: 99%; • Ultrafiltration: 95.91–98.9%; • Microfiltration: 26–56%.	• Removes all dye types; • High separation efficiency; • Low energy requirement; • Relatively gentle operating conditions; • Usually requires smaller space compared to other technologies.	• Concentrated sludge production that would lead to fouling; • High investment and energy costs; • Not suitable for wastewater with a low concentration of contaminants; • Difficulty of produced concentrate disposal or treatment.	[33,39,59,127–129]
Coagulation	Adding coagulant as aggregation aid to increase the sedimentation rate of contaminants	58–100%	• Economically feasible; • Needs simple equipment; • Effective for vat, disperse, and sulfur dyes.	• High sludge production (disposal problem); • Recycling of chemicals is not possible; • Activity relies on pH.	[63,130]
Activated sludge	Utilizes sludge-like bacteria colony to degrade contaminants with aid of aeration	89–95%	• High BOD removal; • Relatively low cost; • Technology already established.	• Time-consuming process; • Unsuitable for non-degradable and toxic compounds; • Possibility of foaming.	[99]
Trickling filter	Spraying wastewater over a filter bed that contains microorganism film on its surface	72–99.8%	• Lower operational cost and area requirements; • Well stabilized sludge.	• Odor emission; • Pumping cost; • Prone to clogging.	[103,104]

Moreover, Table 7 shows other alternative methods that have been tested for treating dye-containing wastewater. Various choices of methods could facilitate the interested parties in considering the treatments for batik or textile wastewater.

Finally, although many organic molecules can be degraded in biological processes, many others are recalcitrant due to their complex chemical structure and synthetic organic origin. Thus, it is important to identify which microorganism is suitable to treat these effluents. Table 8 gives a simple guideline to select the suitable microorganisms to treat batik effluents.

Table 7. Advantages and disadvantages of other physical and chemical alternative methods for dye removal in textile wastewater.

Physical/Chemical Method	Method Description	Advantages	Disadvantages
Photochemical	This process involves the use of a photochemical oxidant, such as hydrogen peroxide or ozone, which is activated by UV or visible light to produce reactive oxygen species (ROS).	No sludge production	Formation of by-products
NaOCl	NaOCl is a strong oxidizing agent that can react with the chromophoric groups of dyes to break down their chemical structure and remove them from the water.	Initiates and accelerates azo-bond cleavage	Release of aromatic amine
Cucurbituril	Cucurbituril is a macrocyclic molecule with a hydrophobic cavity that can selectively bind to and trap guest molecules, including dyes.	Good sorption capacity for various dyes	High cost
Electrochemical destruction	The process relies on the application of an electric current to the contaminated water, which initiates a series of oxidation and reduction reactions that ultimately result in the destruction of the dye molecules.	Breakdown compounds are non-hazardous	High cost of electricity
Peat	Wastewater is pumped through a bed containing peat that allows for dye adsorption.	Good adsorbent due to the cellular structure	Specific surface area for adsorption is lower than activated carbon
Wood chips	The adsorption of dye molecules onto the surface of wood chips, that act as a natural adsorbent material.	Good sorption capacity for acid dyes	Requires long retention times
Silica gels	Silica gel is a synthetic, amorphous form of silicon dioxide with a high surface area and surface reactivity, making it an efficient adsorbent for various pollutants.	Effective for basic dye removal	Side reaction prevents commercial application
Ion exchange	The process relies on the selective affinity of ion exchange resins for specific ions or molecules.	Regeneration; no adsorbent loss	Not effective for all dyes
Irradiation	The process relies on the ability of certain wavelengths of radiation to initiate a series of chemical reactions that ultimately result in the degradation and destruction of the dye molecules.	Effective oxidation at lab scale	Requires a lot of dissolved oxygen

Table 8. Advantages and disadvantages of some of the microorganisms in the biological method of textile effluent treatment.

Microorganism	Method Description	Advantages	Disadvantages
Bacteria (aerobic)	The process relies on the ability of aerobic bacteria to break down and convert the organic pollutants into carbon dioxide, water, and other harmless byproducts.	- Decolorize both azo and anthraquinone dyes; - Production of biogas.	- Low decolorization rates; - Requires specific oxygen catalyzed enzymes; - Requires additional carbon and energy sources.
Bacteria (anaerobic)	The process relies on the ability of anaerobic bacteria to use alternative electron acceptors, such as nitrate, sulfates, or carbon dioxide to metabolize organic pollutants.	- Suitable for large scale application; - Takes place at a neutral pH for sludge treatment system; - Allow obligate and facultative bacteria to reduce azo dyes.	- Generation of toxic substances; - Requires post treatment; - Immobilization and recovery of redox mediator presents a challenge.
Fungi	The process relies on the ability of fungi to secrete enzymes that can degrade organic pollutants, including dyes and pesticides.	- Decolorize anthraquinone and indigo-based dyes at a higher rate.	- Decolorization rate is very low for azo dyes; - Requires special bioreactor and external carbon source; - Needs acidic pH (4.5 to 5) - Inhibition by a mixture of dyes and chemicals in textile effluents.

7. Conclusions

Batik industries pose a threat to the bodies of water and to life inside them if wastewater from the processing is not treated. Dyes, sodium silicate, resin, and wax used in batik production are becoming the main contaminants of the wastewater. This wastewater contains higher BOD, COD, suspended, and dissolved solids, dyes, and heavy metals than the parameters permitted in the environment. Many technologies are available for reducing the terrible impacts of the batik wastewater effluents, although each technology has its own advantages and drawbacks. Despite the limited number of studies available for the application of those technologies for treating batik wastewater, especially at large or commercial scales, this review has summarized the wastewater treatment techniques used in textile industries which have similar characteristics with batik effluents. This review has gathered the latest results of these studies for treating batik effluents to become the general reference for treating the wastewater of batik or textile industries.

The selection of technologies must be made based on the characteristics of the wastewater to be treated, the permitted values released into wastewater, and the available capital, areas, and skilled labor. Most of the time, the application of one technology is not enough to handle all dangerous contaminants in batik factories' wastewater. Combination of two or more technologies may cover the drawbacks of each technology. Although one or more combinations of technologies are suitable to achieve the permitted value for effluents, the cost of investment and operating these technologies might be too high for batik industries, where most of the batik are produced in small scale workshops. Many recent and emerging techniques still need pilot or commercial scale studies to prove that the performance of these technologies is still good at the large scale.

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