Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2022.28028

257 (2022) 185–203
May

A critical review on the treatment of reactive dye wastewater

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Received 17 September 2021; Accepted 3 December 2021

ABSTRACT

Through generations, reactive dyes are known to be one of the most used dyes in the textile industry and are the main source of coloured wastewater. Inarguably, the use of reactive dyes in textile industries has a high probability of bringing potential risk to human health and the environment due to their characteristics being noxious, carcinogenic, and mutagenic. Growing concern about the impact on human life and the environment has led to much research on the treatment of textile wastewater. Throughout the years, research on wastewater treatment has continuously been developed from physico-chemical to biological, electrochemical, conventional, and advanced oxidation methods in which each method offers several advantages and disadvantages in the removal of dyes. However, the existing methods alone are insufficient as textile wastewater may contain a high concentration of chemical oxygen demand and may be difficult to degrade. Due to this, research on further improving the treatment of textile wastewater has constantly been developed. Recently, the use of hybrid or combined treatment processes has raised interest among researchers in the treatment of textile wastewater. It is expected that this technology will be significant in the future, given the latest findings on the removal of reactive dyes from wastewater. This paper presents brief research on reactive dyes and reviews the advantages, disadvantages, and potentials of the current methods employed in the treatment of reactive dye wastewater.

Keywords: Dye removal; Hybrid process; Reactive dye; Textile wastewater; Wastewater treatment

1. Introduction

Throughout the years, the textile industry has been known as one of the largest manufacturing and water-consuming industries in the world. In Malaysia, the growth of the textile industry started in the early 1970s. Man-made textile fibre (batik industry) is highly popular on the East Coast of Peninsular Malaysia and Sarawak, and today, textile industries have contributed to Malaysia's economic growth. In the process of dyeing natural or man-made textile fibres, commonly, a colorant, also known as a dye, is used [1–5]. In the textile industry, reactive dyes are extensively used due to their brilliant colours and advantageous high washing fastness. However, up to 50% of reactive dyes may be lost and discharged as coloured effluent due to the nature of reactive dyes that are difficult to bind with fibres after hydrolyzation [6]. Also, the usage of reactive dyes in textile industries has resulted in the discharge of dye wastewater containing a wide range of non-degradable colourants and toxic compounds such as degradable organics, detergents, de-sizers, dyes, heavy metals, inorganic salts, and stabilising agents [7–14].

The existence of these contaminants in textile wastewater has surely sparked widespread worry because, if not adequately handled, colour and hazardous effluents could be released into water bodies, creating a slew of negative

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Presented at the 5th International Conference on Global Sustainability and Chemical Engineering (ICGSCE 2021), 14–15 September 2021, Virtual Conference organized by Universiti Teknologi Mara (UiTM), Selangor, Malaysia

consequences and finally rendering them unfit for further use [15]. It is reported that, even at low concentrations, the presence of these pollutants would prevent light penetration. This eventually weakens the photosynthetic phenomena in plants, thereby destroying the aesthetic value of water and endangering aquatic life and the environment [16–19]. Moreover, reactive dyes are classified as toxic or carcinogenic agents towards humans, resulting in serious health issues such as allergic dermatitis, skin irritation, typhoid fever, and other diseases [20–24]. Considering the continual concern about the impact on all living organisms and the environment, has led to research on the treatment of textile wastewater [25].

In the treatment of textile wastewater, the effectiveness of the removal of dyes mainly depends on the chromophore as well as the auxochromes of the dyes [15,26]. The chromophore group or conjugated bonds such as carbonyl (–C=O), azo (–N=N–), nitro (–N=O) and quinoid groups are the unsaturated parts of the dye molecules which are responsible for colouration [15,27]. On the other hand, auxochromes are the subsequent group that exists along with azo groups and aromatic rings such as anthraquinone and triphenylmethane within the chromophore group. Auxochromes such as CO, OH, and NH_2 groups are responsible for enhancing the affinity of dyes towards fibres by shifting the wavelength of the light absorbed into the visible region [15,28]. For instance, the colour of anthraquinone and triphenylmethane appears to be blue as it absorbs light in the red or yellow region. This indicates that the colour of dyes is actually the colour of reflected light and that it differs depending on the type of auxochromes present [15].

To date, physico-chemical, biological, electrochemical, conventional, and advanced oxidation methods have been utilised in the treatment of textile wastewater, each with its own set of advantages and limitations in the removal of dyes. Reactive dyes have been known to be the most problematic class of dyes as it has always been a challenge to assess the appropriate treatment for the removal of reactive dyes from wastewater due to their poor degradability and complex reactive dye structure [27,29]. Thus, a depth analysis is required to obtain satisfactory removal of reactive dyes. This paper aims to present brief research on reactive dyes and to review the advantages, disadvantages, and potentials of the current methods employed in the treatment of reactive dye wastewater.

2. Classification of dyes

The discharge of effluent or wastewater has always posed grave problems for the ecosystem and human development [30]. Colouration is thought to be responsible for 17% to 20% of water pollution [27]. Colouration occurs due to the presence of chromophore groups or conjugated bonds, which are the unsaturated parts of the dye molecules [15,28]. The main causes of water pollution are food, painting, printing, and plastic industries, as well as textile industries [31]. Globally, the textile and dyeing industries are known to be the major causes of water pollution due to the considerable amounts of dyes and wastewater generated during the process, as well as other negative environmental impacts [20]. In general, dyes can be classified into acid dyes, dispersed dyes, basic dyes, direct dyes, reactive dyes, sulphur dyes, and vat dyes depending on their applications, solubility, and chemical structures [32]. Among various classes of dyes, reactive dyes are the most consumed dyes in textile industries, which represents 25% of the world's total dye market [1,20,33].

In brief, reactive dyes are anionic and water-soluble dyes that exhibit a wide range of chemical structures, primarily based on substituted aromatic and heterocyclic groups [20,33]. They can be formed by forming covalent bonds between reactive groups and hydroxyl or amino groups found in the textile fibres [28,32,34]. Reactive dyes are commonly used in the textile industry due to their ability to produce brilliant colours and their advantageous high washing fastness [15,20,26]. Despite that, reactive dyes are identified as the most problematic class of dyes due to their acidic properties and poor nature in binding with fibres after hydrolysation [28]. They are also known to be difficult to degrade as colour may still remain in the effluent after the treatment process [20].

The use of reactive dyes in the textile industry has resulted in the discharge of dye wastewater that is rich in colour, pH, temperature, and turbidity, as well as a high chemical oxygen demand (COD) that is extremely visible, even in traces (approximately 50 to 18,000 mg/L) [35–37]. This has caused a major concern in developing countries as the presence of dyes, even at low concentration, will destroy the aesthetic value of water, which undoubtedly will deteriorate the environment as they are discharged into water bodies [16–19]. They could also endanger aquatic life and cause serious health problems in humans [20–24]. Concern about the impact on aquatic life, human life and the environment has led to research into the treatment of textile wastewater [25].

3. Treatment of textile wastewater

Nowadays, the removal of reactive dyes from textile industries has not only become a major environmental concern but also a challenge [32]. The textile industry involves several stages that use huge quantities of water, generating a large amount of wastewater. The wastewater discharged by the industry is often rich in colour, pH, temperature, turbidity, and has a high COD (approximately 50–18,000 mg/L) and has always posed a grave problem for human health and the environment [4,12,38]. Therefore, identifying an appropriate approach for the removal of reactive dyes from textile wastewater before discharging it into water bodies would greatly benefit human health and the environment [3,32].

In the treatment of textile wastewater, the effectiveness of the removal of dyes mainly depends on the chromophore as well as the auxochromes of the dye [15,26]. To date, there are numerous methods that have been employed in the treatment of textile wastewater, including physico-chemical, biological, electrochemical, conventional, and advanced oxidation methods, whereby each treatment process provides its own specific characteristics. Although there are various treatment processes that have been introduced, not all of the methods are suitable to be practised due to their disadvantages. According to [32], an ideal wastewater treatment method would be one that is able to efficiently remove large amounts of pollutants from wastewater in a short period of time without generating more hazardous by-products. In this section, the potential of current methods employed in the treatment of reactive dye wastewater, as well as their advantages and disadvantages, will be discussed.

3.1. Biological processes

A biological process, which can be used to remove nutrients, solid and organic matter, and decolourise a wide range of dyes from wastewater, is one of the alternatives to treating textile wastewater [31,39]. In general, the biological approach incorporates the use of several microbial sources such as algae, yeasts, fungi, actinomycetes, and bacteria. This approach is known to be adept at removing dyes from large amounts of wastewater. It is also believed that this method is the cheapest and safest when it comes to the removal of dyes [31,32]. Normally, during the process, the microorganisms used will metabolise and synthesise enzymes that help in breaking down biodegradable organic matter into simpler substances such as carbon dioxide, energy, and water, which later will function for their growth and reproduction [25,31,39–41].

Many efforts have been made in the process of decolourising wide ranges of dyes using a biological approach, which includes the use of varieties of bacteria such as *Bacillus cereus*, *Escherichia coli* and *Pseudomonas pseudomallei* [25,31,39–41]. Due to their ability to survive in harsh conditions, extremophilic microorganisms are commonly used for the treatment of textile wastewater. Among the potential microorganisms that have been demonstrated in the removal of dyes are *Aspergillus Niger*, *Bacillus cereus*, and *Chlorella* sp. [25]. The use of rhizobacteria has also shown great potential in the process of decolourising various textile dyes [41]. In a biological approach, the study on the ability of decolourisation of dyes is conducted by growing a culture of the type of strain in a medium and the influence of environmental effects such as pH, temperature, and initial dye concentration will be evaluated. In essence, the effectiveness of the treatment process depends on the adaptability and microorganism activities under optimum conditions.

In brief, the applications of biological treatment processes mainly involve the use of conventional treatment methods such as activated sludge, lagoons, and trickling filters. However, these methods alone are inefficient for colour and pollutants removal. This has led researchers to shift the use of conventional biological approaches to advanced bioremediation processes or combination processes [40,41]. According to [42], better treatment efficiency can be achieved by combining biological methods with other methods. Studies have also been conducted on the decolourisation of dyes from textile wastewater by emerging new bacteria. Despite the efforts made by researchers, the biological approach is still limited to removing colour rather than pollutants. Table 1 shows examples of applications for the use of yeast, fungal, and bacterial cultures or strains in textile wastewater.

According to Table 1 from literature [43–48], biological processes can treat reactive dyes in textile wastewater. A study from [46] showed that the use of fungal culture, namely *Rhizopus arrhizus*, is able to decolourise reactive dyes up to 100%. However, the application is limited as the decolourisation process may require days to complete. Another study [31] shows that the use of bacterial culture, namely *Lactobacillus delbrueckii*, is able to decolourise dyes with an initial concentration of 100 mg/L in a shorter period of time compared to that of fungal culture. The use of yeast culture has also been successfully applied, and maximum colour removal is attained at a higher initial dye concentration, 300 mg/L, within a shorter period of time compared to fungal and bacterial culture [43].

However, this method is limited as it requires a large operational area, a certain series of steps, and a certain period for the process of culturing and isolating culture from

Table 1
Applications of biological treatment processes in textile wastewater

Strain	Dye	Concentration (mg/L)	% Decolourisation	Time	References
		Yeast culture			
Trichosporon akiyoshidainum HP 2023	Reactive Black 5	300	100	24 h	[43]
Candida krusei	Basic Violet 3	10	100	24 h	[44]
		Fungal culture			
Aspergillus fumigatus	Methylene Blue	12	93.43	120 min	[45]
Rhizopus arrhizus	Reactive Red RB	100	71.83	8 d	[46]
	Reactive Black B	100	100	3 d	
	Remazol Blue	100	100	2 d	
	Methylene Blue	100	92.5	8 d	
		Bacterial culture			
Halomonas	Reactive Black 5	50	87	5 d	[47]
	Reactive Red 152	50	85	5 d	
Lactobacillus delbrueckii	Reactive Black 5	100	55	48 h	[48]
	Reactive Orange 16	100	63	48 h	

collected samples. Also, the treatment process may take a longer period of time depending on the type of strain or culture, concentration, and type of reactive dyes. As the growth of the textile industry keeps on increasing every year, the use of biological treatment processes for textile wastewater treatment becomes uneconomically feasible. This is due to the slow culture growth rate, low biodegradability, and slow reaction time in degrading reactive dyes. This eventually raised concerns among the public as it may cause irresponsible parties to discharge untreated textile effluent into water bodies. The incomplete destruction of organic matters and limitations through biological approaches have caused the treatment of reactive dye wastewater to shift to the use of physico-chemical processes.

3.2. Physico-chemical processes

Physico-chemical processes are known to be a straightforward method that is commonly accomplished through mass transfer mechanisms. Compared to biological and chemical oxidation processes, physico-chemical processes are the most commonly used method due to their simplicity and efficiency in dye removal. Examples of physico-chemical processes are coagulation–flocculation, adsorption, and membrane filtration [32]. The applications of these methods to textile wastewater are tabulated in Table 2 along with their decolourisation efficiency.

3.2.1. Coagulation–flocculation

Coagulation–flocculation is a chemical treatment that is proven to be highly effective in reducing COD and a significant amount of colour (by 70% to 80%) in wastewater. This process can be divided into three main phases, which are coagulation, flocculation, and sedimentation. In the first phase, the coagulation process takes place using coagulants, in which the amount of coagulants used mainly depends on the concentration of dyes and type of effluent [15,25]. In industries, coagulants from inorganic salts such as aluminium sulphate ($Al_2(SO_4)^3$), ferric chloride ($FeCl_3$), polyaluminium chloride (PAC) and synthetic organic polymers are commonly used [25]. During coagulation, electrostatic attraction takes place between oppositely charged soluble dye and polymer molecules whereby the coagulants used will help in destabilising colloidal particles, soluble compounds, and very fine solid suspensions [15,25]. The process is then followed by flocculation, which is a physical process that allows flocs to form by combining small particles into larger aggregates for easier separation [49]. Lastly, sedimentation takes place, in which the compounds or particles in the form of floc are removed from the process. Recently, in enhancing the treatment of effluent discharged by textile industries, the use of the coagulation method has shifted to combining with other treatment processes such as biological processes.

3.2.2. Adsorption

Among physico-chemical processes, adsorption has received wide attention due to its vast advantages in removing dyes from textile wastewater [29,32,50,51]. Compared to other treatment processes, adsorption is known to be an ideal process as it generates higher treated water quality [32]. Adsorption is a mass transfer process involving two interfacial layers (liquid-liquid, liquid-gas, gas-solid, or solid-liquid interface). The process may take place through chemical sorption (chemisorption) and physical sorption (physisorption). For physisorption, the reaction takes place through the Van der Waals force, while chemisorption takes place *via* chemical bonding between the adsorbent and the adsorbate. Within the process, the substance that is being adsorbed is known as an adsorbate, while the adsorbing material is known as an adsorbent [25,32,52]. The capability of adsorbent regeneration and effectiveness of dye removal mainly depends on the selection of an adsorbent [29,51].

In adsorption, diffusion occurs in which the dye molecules from the wastewater are adsorbed on the surface of the adsorbent. Within the process, molecular interaction occurs between the dye molecules and the adsorbent, involving either monolayer or multilayer adsorption, depending on the type of adsorbent used. As the process takes place, the adsorbent will be filtered from the process for removal. In essence, the efficiency of the adsorption process is highly dependent on the physical and chemical properties of the adsorbents, such as contact time, pH, particle size, temperature, and surface area. Historically, one of the most commercial and versatile adsorbents is activated carbon from coal, which is highly effective in removing a wide range of dyes due to its large surface area and highly porous structure [50,53]. However, the commercial use of activated carbon as an adsorbent is hampered by its high operational cost and regeneration limitations [29,51,54]. Consequently, for economical and practicable applications, numerous attempts have been made by researchers to search for an alternative adsorbent from waste resources or biomass materials such as coffee husks [18], corn husks [55], coconut shells [56] and exhausted coffee waste [57].

3.2.3. Membrane filtration

The potential for rejecting large amounts of pollutants from wastewater while producing high-quality treated water has led to an increase in the use of membrane technology [24]. Membrane filtration is a physical process that is widely used due to its simplicity and ease of replacement [5]. Compared to traditional physico-chemical processes, membrane technology requires preliminary chemical treatment, followed by physical means, which is provided by the membrane [58]. It is commonly used in industries to treat secondary and tertiary municipal wastewater. The term "membrane" refers to a permeable or semi-permeable phase that takes place by forming a barrier that allows certain substances to permeate while hindering foreign substances (retentate) [25,59]. In the treatment of textile wastewater, dye molecules from the wastewater will diffuse through the pores of the membranes. The transfer of dye molecules is assisted by driving forces such as energy, concentration, pressure, and temperature gradients [60]. In meeting the discharged limits and retaining the cleanliness of water, the membrane helps in separating fluids, suspended or dissolved, colloidal from wastewater [25,61].

Table 2
Applications of physico-chemical processes in textile wastewater

Process	Dye	Operating conditions	% Removal	References
Coagulation–flocculation	Reactive dyes	- Using alum at a conc. of 6,000 mg/L and pH 2.4 - Using $MgCl_2$ at a conc. of 4,000 mg/L and pH 10.4 - Using PAC at a conc. of 2,000 mg/L and pH 4.1	Colour = 90 Colour = 99 Colour = 100	[63]
	Dye effluent	- Using 450 mg/L of $FeSO_4$ as a coagulant at pH 12 - Using 500 mg/L of $FeCl_3$ as a coagulant at pH 12	Colour = 92, COD = 62 Colour = 91, COD = 64	[64]
	Reactive dyes	- Using alum at pH 3.8 to 4.6 - Using $MgCl_2$ at pH 10.4 to 10.9 - Using PAC at pH 3.9 to 5.2	Polyaluminium chloride was most effective where: Colour > 99 COD = 90.6 to 96.3	[26]
Adsorption	Crystal Violet	Using waste coffee husks (WCH) as an adsorbent at 25°C, pH 3, and a dosage of 1.5 g	Adsorption = 98	[18]
	Methyl Orange	Using thermally chemically activated eggshell as an adsorbent at a conc. of 12.5 mg/L, 20 min. contact time and a dosage of 2 g	Adsorption = 98.8	[65]
Membrane filtration	C.I. Reactive Blue 38	Using microfiltration membrane at conc. of 10 g/L, pH 7 and pressure of 3 kPa	Dye rejection = 91	[59]
	Reactive Black 5 Reactive Blue 15 Reactive Orange 16 Reactive Yellow 145 Reactive Red 194	Using nanomembrane at conc. 16 mg/L, 60 min. and pH 6.4 to 7.1	Dye rejection = 90 to 97 COD = Reduced to zero	[5]

In the past, many researchers have studied the use of membranes in treating textile wastewater. The application of membranes in the industry has been presented in the form of nanofiltration (NF), ultrafiltration (UF) and reverse osmosis (RO) and has been proven to be potentially effective in reducing biochemical oxygen demand (BOD), COD, and colour from textile wastewater [25,29,61]. The separation characteristics of a membrane mainly depend on the particle size and membrane pore diameter [62]. Nevertheless, in terms of economics, it is undeniable that the operating cost will be high as the life span of a membrane is usually short as it can be critically challenging to prevent a membrane from fouling [58]. This has led researchers to modify or combine membrane technology with other treatment processes such as coagulation–flocculation, adsorption, and electrochemical processes to reduce membrane fouling, operating costs, and enhance the removal of dyes from wastewater [24,42].

It is best known that biological approaches alone cannot completely degrade dyes from wastewater, and this has led researchers to shift to the use of physico-chemical processes. In reference to Table 2, studies from [5,18,26,59,63–65] show that physico-chemical processes are promising in treating reactive dyes from textile wastewater. For coagulation–flocculation, the percentage of colour removal depends on the pH of the wastewater and the type and dosage of coagulant used. The effectiveness of the process is reviewed in a study [63], which proves that the decolourisation of dyes can be achieved up to 100% using PAC at a concentration of 2,000 mg/L and pH of 4.1. However, this process is mainly effective for dispersed and sulphur dyes but ineffective for water-soluble dyes such as reactive dyes, as it only coagulates and does not settle. Also, the use of this method is uneconomical as it requires pH adjustment, high-cost chemical reagents, and generates large amounts of sludge and total dissolved solids, making it expensive and complicated.

On the other hand, the adsorption method is highly effective in removing dyes from textile wastewater. Like in biological approaches, adsorption can be carried out several times until the adsorbent is spent. In adsorption, the removal percentage is highly dependent on the type and amount of adsorbent used. As the use of activated carbon from coal is expensive, the commercial application of the adsorbent is hindered. This issue is later overcome by using low-cost materials as adsorbents. A study [65] has reported that the use of low-cost adsorbent from thermally chemically activated eggshell is able to remove colour up to 98.8%. Despite the efforts made by researchers on the use of low-cost materials as an adsorbent, the applications of these adsorbents are limited as they may be difficult to regenerate, and sometimes the loss of an adsorbent may lead to colourisation of water. Also, the spent adsorbent may be hazardous, making it difficult to handle. This makes the adsorption method expensive as a high operational cost is required.

As for membrane filtration, the amount of dye to be adsorbed on the surface of the membrane depends on the size of the dye particle and the pore diameter of the membrane. Research carried out by [5] shows that the use of nanomembrane is able to treat various types of reactive dyes at conc. 16 mg/L, 60 min, and pH 6.4 to 7.1 with 90%–97% dye rejection and zero COD. This proves the statement that membrane filtration is effective in removing all types of dyes [15]. Despite that, there is a high tendency for membrane fouling, which leads to a shorter membrane life and lower productivity for dye removal over time. This causes a need for high maintenance costs as the membrane must be changed over time to ensure continuous usage, thus making it limited in the treatment of wastewater.

In summary, physico-chemical processes are effective and economical only when the solute concentrations in textile wastewater are high. The use of these methods alone is inefficient as ultimate removal or complete degradation of dyes in textile wastewater could not be achieved as they only focus on the transfer of contaminants. Also, these methods are limited to small volumes of textile wastewater and are only capable of the removal of certain types of dyes. This has led researchers to develop the use of conventional chemical oxidation processes in treating textile wastewater.

3.3. Chemical oxidation processes

As reactive dyes are difficult to degrade through physico-chemical or biological processes, chemical oxidation processes are introduced. Chemical oxidation is widely used in industries and seems to hold potential for future use in the treatment of textile wastewater due to its effectiveness and simplicity in the degradation of dyes, besides inhibiting microbial growth and preventing the formation of toxic inorganic compounds [15,33,51,66]. The method involves the use of an oxidising agent that helps in the generation of hydroxyl radicals ($OH^{\bullet}$), which later effectively mineralise organic compounds into carbon dioxide, mineral acids and water [33]. Basically, chemical oxidation processes can be divided into conventional oxidation and advanced oxidation processes. These oxidation processes can either be used individually or in synergism with one another [29].

3.4. Conventional oxidation processes

There are various techniques that can be categorised under conventional chemical oxidation processes, which include the usage of oxidising agents like O_3, H_2O_2, and Cl compounds. O_3 and H_2O_2 are known to form strong non-selective hydroxyl radicals at high pH values that will effectively breakdown the complex aromatic rings of dyes, and the small formation of non-chromophore molecules will further help in reducing the effluent colour [29,51]. Contrarily, the use of Cl compounds is less effective in the removal of reactive dyes from textile wastewater. Table 3 shows the applications of conventional chemical oxidation processes in treating textile wastewater.

3.4.1. Ozonation

Ozonation (O_3) is a powerful oxidising agent compared to other oxidising agents such as chlorine and H_2O_2 that act as potential alternatives in decolourising textile wastewater. Similar to H_2O_2, it forms a strong, non-selective hydroxyl radical that works best at an alkaline pH [64,67–70]. It is considered an environmentally friendly method as it may

Table 3
Applications of conventional oxidation processes in treating textile wastewater

Process	Dye	Operating conditions	% Removal	References
Ozonation	Reactive Black 5	- Initial conc. of 40 mg/L and 100 mg/L at an incubation time of 150 and 300 min, respectively - Ozone output of 1.16, 3.81, 18.79, and 40.88 mg/min is used at an incubation time of 2 h - Flow rate of gas stream of 0.6, 1.4, 2 and 4 LPM is used	Colour = 96.9 COD = 77.5	[12]
	Reactive dyes	Temperature is set at 35°C and the reaction time 6 h	Colour = 92.2	[68]
	Acid Red 337 Reactive Orange 16	An ozone dosage of 12 mg/L min is applied at pH 7 for 30 min	Colour = 60 to 91 COD = 9 to 17	[69]
	Dye bath effluent	Oxygen with a purity of 99.5% is used and operated at pH 6.2 for 25 min	Colour = 58	[6]
Hydrogen peroxide	Crystal Violet	H_2O_2 dose of 2 mL/L is used	Colour = 97	[74]
	C.I. Basic Blue 9	Use a dosage of 10 g/L of H_2O_2 at pH 12	Colour = 95	[67]
Chlorine	Red and blue dyes		Colour = 96 to 98 Turbidity = 71 to 72	[13]

eliminate contaminants, coloured substances, odours or even bio-recalcitrant chemical species that are resistant to biological processes without creating harmful or significant by-products. In general, ozone can react with chemicals either in direct or indirect ways. In the direct oxidation method, the dipolar structure of ozone molecules will attack the unsaturated bond in the dye. This method is known to be selective as only certain parts of organic compounds are degraded. Meanwhile, the indirect oxidation method operates in high media whereby hydroxyl radicals generated decompose organic compounds into carbon dioxide and water [71]. In general, ozone is highly soluble in water and easily breaks down into free radicals, such as hydroxyl ($OH^{\bullet}$), $HO_3^{\bullet}$, $HO_4^{\bullet}$ and superoxide O_2^{-}[25,64,69].

In the treatment of textile wastewater, dyes lose their colour by breaking down the functional groups or conjugated C–C bonds in the dye chromophores. This can be further explained by the fact that during ozonation, the free hydroxyl radicals formed will attack the bonds and later shift the wavelength of the light absorbed onto the dye molecules from their visible region [69]. However, this method has a short life span and requires pH adjustment, making it difficult to handle and costly [29]. In providing economy and effectiveness in treating textile wastewater, some extra modifications should be applied to the ozonation process, such as combining ozone with biological processes, hydrogen peroxide, UV radiation, or solid catalysts [71].

3.4.2. Hydrogen peroxide

Another oxidising chemical agent that is often used in the process of decolourisation of wastewater is hydrogen peroxide (H_2O_2). Similar to O_3, it forms a strong, non-selective hydroxyl radical that works best at an alkaline pH [71]. This method has high redox potential, allowing it to be effective against organic molecules such as dyes [66]. According to research, the use of H_2O_2 as an oxidising agent in wastewater is extremely effective because it is capable of reducing or eliminating complex dyes and the conjugated C–C bonds in the dye chromophores [29,72]. It decomposes exothermically into water and oxygen gas, depending on the pH of the solution, temperature, and presence of impurities in the effluent [73]. However, this method has a low degradation rate compared to O_3 as fewer hydroxyl radicals are generated during the process [29]. Thus, over the years, to improve the effectiveness of colour removal from wastewater, various combination techniques, such as combining H_2O_2 with UV rays or O_3, have been studied [74,75].

3.4.3. Chlorine compounds

Chlorine is a traditional chemical oxidation method that is widely used in wastewater treatment and is available in liquid form (as sodium hypochlorite, NaOCl), gaseous form (as Cl_2), or solid form (as calcium hypochlorite, $Ca(OCl)_2$) [66,73]. Basically, the amino groups present within the molecular structure of dyes are the easiest to attack by chlorine. Precisely for this reason, chlorine compounds are capable of removing colour from wastewater by attacking the amine group and also help in accelerating the destruction of nitrogenous bridges within dye molecules. Over the years, the use of chlorine in the decolourisation of acids and direct dyes has been widely reported, whereby the rate of effectiveness of chlorine increases with the amount of chlorine added with decreasing pH. However, during the process, chlorinated by-products such as metal complexes may be formed, thus making it less efficient for chlorine compounds to treat reactive dye wastewater [66]. The concern over the use of chlorine compounds has led to the use of other chemical oxidants such as O_3 and H_2O_2 [73].

In reference to Table 3, past studies [6,12,13,67–69,74] have shown that the use of oxidising agents like O_3, H_2O_2 and Cl compounds is capable of treating textile wastewater. For ozonation, it can be used in a gaseous state and would not increase the volume of wastewater or lead to sludge generation. Studies from [12,69] reveal that the COD removal is lower than colour removal in which factors such as O_3 concentration, pH, and incubation time will affect the colour and COD removals. The highest removal of colour and COD was achieved by 96.9% (using 40.88 mg/min at 300 min incubation time) and 77.5% (using 40 mg/L of RB5 initial concentration at 120 min incubation time) [12]. The low COD removal is attributed to the structure of dye molecules that are oxidised into smaller molecules such as aldehydes and ketones, which consist of a considerable level of COD. Although ozonation has the potential to remove reactive dyes from textile wastewater, the process may be complex and costly as it requires the use of an electrical supply to ensure a continuous oxygen supply. Apart from that, the use of O_3 may also be expensive as it requires post-treatment for complete dye degradation and for the removal of by-products generated from the process.

Meanwhile, for hydrogen peroxide, the removal efficiency depends on the pH and concentration of H_2O_2 used. [67] evaluated the colour and COD removals for C.I. Basic Blue 9 dye using H_2O_2 and it was found that the process is capable of removing 95% of colour and 65% of COD at pH 12 with 10 g/L of H_2O_2. However, it may be difficult to store H_2O_2 and the addition of H_2O_2 into wastewater may be costly. Also, the use of H_2O_2 alone is limited as fewer hydroxyl radicals are produced in the process, making it inefficient in completely degrading dyes from textile wastewater. On the other hand, chlorination is cheap and easily available for treating wastewater. A study [13] shows that this method is able to remove COD and turbidity in textile wastewater by 98% and 72%, respectively, depending on the type of dyes in the wastewater. Despite that, the use of chlorine is limited to certain types of pollutants and is difficult to handle. It also has low biodegradability and may lead to the transformation of pollutants into carcinogenic agents.

Although the use of conventional oxidation processes can treat textile wastewater, it is shown that they are incapable of removing pollutants completely from textile wastewater due to the low degradation rate, which is equated to the limited generation of hydroxyl radicals. Additionally, the addition of the oxidising agents may be costly, and the sludge produced from the processes may be harmful to the environment and may lead to disposal problems. Compared to other treatment methods, these processes are dangerous to transport and store. Thus, in improving the effectiveness of removing microbial loads and reducing the cost of oxidising agents used, further improvements are made by introducing advanced chemical oxidation processes.

3.5. Advanced oxidation processes

Previously, conventional oxidation processes have been found to be difficult in oxidising dyes or complex organic compounds due to the low biodegradability of the processes [76]. This has led to the study of advanced oxidation processes (AOP), which seem to hold the potential to effectively remove pollutants from wastewater [6]. Like conventional oxidation processes, they are a chemical treatment method that requires the use of oxidising agents. Compared to conventional oxidation processes, they are very active and have oxidising agents that hold faster oxidation rates due to the adequate amount of hydroxyl radicals ($OH^•$) generated by the processes [6,29,51,71]. These processes are widely used for the degradation of dyes due to their ease of application in eliminating non-degradable organic components and avoiding the removal of residual deposits [77,78].

In general, AOP can be operated either through the means of acoustic cavitation (ultrasonic radiation) or *via* photocatalytic oxidation (a semiconductor catalyst that is activated through sunlight) or *via* Fenton reaction (which involves the reaction between Fe ions and H_2O_2) [29]. The mechanism of AOP can be divided into two steps: (1) hydroxyl radicals ($OH^•$) will be generated either photochemically or non-photochemically; and (2) oxidation by radicals formed whereby pollutants (such as dyes) are converted into carbon dioxide and water [79]. AOP applications include the Fenton reaction, O_3, H_2O_2 and UV light, as well as their combinations, such as O_3/H_2O_2, O_3/UV, UV/ H_2O_2 [6]. Table 4 shows the applications of advanced chemical oxidation processes in treating textile wastewater.

3.5.1. Ozone-based

As conventional ozonation processes are ineffective in removing pollutants from wastewater, extra modifications are applied to the ozonation process, such as combining O_3 with H_2O_2, UV radiation, or solid catalyst. It is expected that the ozone-based methods will provide a more economic and effective treatment of textile wastewater [71]. Combination methods involving O_3 have been exploited and, unlike the normal ozonation methods, ozone-based methods form $OH^•$ radicals that tend to attack ionic, neutral, and dissociated organic substances through higher redox potentials, helping to further increase the removal of colour from wastewater in industries [66,80]. According to [70], ozone-based methods may decolourise all types of dyes except for vat dyes and non-soluble dispersed dyes, whereby the applications of ozone-based methods depend on the initial dye concentration and the amount of O_3 added.

In the process of combining ozone and hydrogen peroxide (O_3/H_2O_2), also known as Peroxone, the decomposition rate of O_3 into hydroxyl radicals is improved due to the addition of H_2O_2 into the ozonation process at high pH [71,75,76]. The reaction that takes place can be summarised in Eq. (1).

$$H_2O_2 + 2O_3 \rightarrow 2OH^• + 3O_2 \quad (1)$$

Despite the efforts made by combining O_3 with H_2O_2, the process is still considerably inefficient for COD reduction compared to colour removal. As for the combination of ozone and UV rays (O_3/UV), all kinds of UV light sources are used to further enhance the generation of $OH^•$ radicals to break down the functional groups or conjugated C–C bonds in the dye chromophores [80]. Compared to conventional ozonation methods, this combination method is capable of decomposing organic compounds completely into carbon dioxide and water [70]. Eqs. (2)–(4) depict the reaction occurring within the O_3/UV process.

$$O_3 + h\nu + H_2O \rightarrow H_2O_2 + O_2 \quad (2)$$

Table 4
Applications of advanced oxidation processes in treating textile wastewater

Process	Dye	Operating conditions	% Removal	References
Ozone-based (O_3/H_2O_2)	Dye effluent	Using 5 mg/L of peroxide at 23°C for 15 min	Colour = 99 COD = 54	[64]
	Acid Blue 92	Using 120 g O_3/h of ozonised air for 1 h at 23°C	COD = 80	[71]
Ozone-based (O_3/UV)	C.I. Reactive Blue 19	Operate at pH 6.2, 25°C and dye conc. of 800 mg/L for 90 min	COD = 57 TOC = 27	[78]
Hydrogen peroxide-based (UV/H_2O_2)	Reactive Blue 19	Using a dosage of 2.5 mmol/L of H_2O_2 at pH 3 and 55 watts for 30 min	Colour = 99	[83]
	Dye bath effluent	Run for 81 min at pH 6 with an initial conc. of 0.6 M H_2O_2	Colour = 98.77 COD = 86.11	[84]
Fenton reaction	Dye effluent	pH is adjusted to 3 where a conc. of 200 mg/L of $FeSO_4$ and $FeCl_3$ are added, together with 400 mg/L of H_2O_2	For $FeSO_4$ Colour = 95, COD = 78 For $FeCl_3$ Colour = 71, COD = 64	[64]
	Acid Red 337 Reactive Orange 16	Suitable ratios of dosage of Fe(II) to H_2O_2 are used where 300 mg/L of H_2O_2 and 250 mg/L of $FeSO_4$ at pH 3	Colour = 99 COD = 82	[69]
	Disperse Red 167	At 25°C , run for 100 min. with an initial dye conc. of 100 mg/L and a pH of 3	Colour = 77.19, COD = 78.03	[82]
	Disperse Yellow 23		Colour = 84.66, COD = 75.81	
	Disperse Blue 2BLN		Colour = 79.63, COD = 78.14	
	Acid Basic Direct Disperse Reactive	All dyes are mixed in equal ratios and operated at pH level below 3.5	Colour = 97 COD = 90	[85]

$$H_2O_2 + h\nu \rightarrow 2OH^{\bullet} \quad (3)$$

$$2O_3 + H_2O_2 \rightarrow 2OH^{\bullet} + 3O_2 \quad (4)$$

3.5.2. Hydrogen peroxide-based

The use of hydrogen peroxide alone is expensive and is ineffective in treating textile wastewater, both in acidic and alkaline conditions [75]. This has led to modifications or improvements in the use of H_2O_2 in wastewater industries by combining it with other methods such as UV radiation. The use of UV rays in the presence of H_2O_2 (direct photolysis of H_2O_2) is known to be capable of destroying the chromophore structure of dyes. Precisely for this reason, the H_2O_2/UV process is widely used in the treatment of textile wastewater [73]. Normally, during the process, H_2O_2 in wastewater will break down to form radicals, ions, or smaller particles that produce HO_2^- [76]. HO_2^- is the acid-base equilibrium in H_2O_2, which is responsible for absorbing UV rays to form highly powerful $OH^{\bullet}$ radicals [66,75]. The generation of these $OH^{\bullet}$ radicals will later lead to the degradation of dyes into carbon dioxide and water [29], [51].

In brief, the use of UV rays is advantageous as it acts as an energy dissipating component to activate the decomposition of H_2O_2 to generate a high concentration of $OH^{\bullet}$ radicals. Compared to other treatment processes, the use of H_2O_2/UV offers a few advantages, such as less treatment time, the ability to operate in ambient conditions, and no sludge generation [29,51,75]. Despite that, the application of the H_2O_2/UV process is limited to certain types of dyes due to its low oxidation potential [15]. In addition, the use of UV rays during the process may lead to higher energy costs [29]. In the removal of dyes, the effectiveness of the process is mainly influenced by parameters such as pH, dye molecular structure, and the intensity of the UV radiation [29,51]. Additionally, the concentration of H_2O_2 added into the process also influences the removal of dyes. Thus, it is crucial to limit and optimise the addition of H_2O_2 as an excessive amount of H_2O_2 may lead to scavenging effects that later reduce the oxidation rate of H_2O_2 to form $OH^{\bullet}$ radicals. Conversely, a low amount of H_2O_2 may limit the generation of $OH^•$ radicals and eventually decrease the oxidation rate [79].

3.5.3. Fenton reaction

The Fenton process involves the combination of Fe^{2+} and H_2O_2 that is effective in degrading various types of organic compounds, including both soluble and insoluble dyes from wastewater. It is known as one of the oldest AOPs and is resistant to biological degradation. In general, this approach is known to be adept at generating $OH^•$ radicals and is highly reactive in attacking dyes compared to conventional treatment processes such as coagulation–flocculation and biological processes [25,29,75]. Also, compared to other oxidation processes such as the UV/ H_2O_2 process, the Fenton process is cheaper, simpler to operate, has no energy consumption, has a fast reaction time, and is able to work at various temperatures [79,81]. In short, the study on the removal of dyes is influenced by the concentration of ferrous ions, the concentration of H_2O_2, the pH of the solution, and the initial concentration of dyes. According to [75], the decolourisation of dyes increases as the concentration of ferrous ions increases due to the high oxidation potential of $OH^•$ radicals generated. This has led many textile industries to apply the Fenton process in the treatment of textile wastewater. However, the $OH^•$ radicals generated may be limited in their operation as they have a very short life span.

In brief, the mechanism of the Fenton process involves the oxidation and coagulation of organic compounds. Complex organic pollutants will be oxidised during the process by promoting the decomposition of H_2O_2 [29,64,69]. Normally, in reducing colour and COD from textile wastewater, organic matter will react with H_2O_2 in the presence of ferrous (Fe^{2+}) ions under acidic conditions [25,69,82]. It is suggested that the process be run in an acidic environment to avoid the formation of ferric hydroxo complexes and the decomposition of H_2O_2 [64]. As Fe^{2+} ions are oxidised by H_2O_2, $OH^•$ radicals will be formed and eventually attack the dye molecules, thus destroying the chromophore and chromogen of the dye molecules [69]. The reaction that takes place in the Fenton process can be summarised in Eq. (5).

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^• + OH^- \quad (5)$$

In the case when the concentration of Fe^{2+} is high or in the absence of a substrate, the $OH^•$ radicals formed from the previous reaction can further oxidise the Fe^{2+} generated into Fe^{3+}, as shown in Eq. (6) [81]. Normally, the formation of Fe^{3+} will result in the formation of coagulant, and the precipitation of $Fe(OH)_3$ which will aid in the removal of suspended solids from wastewater [82].

$$Fe^{3+} + OH^• \rightarrow Fe^{3+} + OH^- \quad (6)$$

In reference to Table 4, as the existing technologies are inefficient enough, researchers have introduced the usage of advanced chemical oxidation processes (AOP) as a potential alternative in decolourising dyes and reducing recalcitrant toxicants from wastewater. In AOP, the decomposition of organic compounds from wastewater is highly dependent on the $OH^•$ radicals. The $OH^•$ radicals generated are highly reactive and are capable of reacting with most dyes at a high reaction rate. One of the AOP methods is ozone-based methods such as combining O_3 with H_2O_2 or UV radiation. These methods have been exploited by many industries due to their effectiveness in attacking ionic, neutral, and dissociated organic substances through their higher redox potential. For O_3/H_2O_2, the effectiveness can be reviewed by a study [64] in which 99% of colour and 54% of COD were removed from the process using direct ozonation at 23°C for 15 min. This method has been proven to be significantly effective in attacking bonds or aromatic rings of dyes in the presence of $OH^•$ radicles, in which the COD removal increases with the intensified reaction time due to the reaction between O_3 and H_2O_2. Meanwhile, for the use of O_3/UV, there is not much difference in colour removal, but it greatly affects the COD and total organic carbon (TOC) removals by 57% and 27%, respectively, at a dye concentration of 800 mg/L, pH of 6.2 and 25°C for 90 min [78]. However, as the concentration of initial dyes increases, more amounts of O_3 are likely to be consumed within the process to enhance mass transfer and ensure the high removal of colour from wastewater is achieved. This makes ozone-based methods expensive as O_3 has a short life span and the process requires a large amount of O_3. Additionally, despite having a high removal of colour, the COD and TOC removals are limited.

In another study involving the use of UV rays in the presence of H_2O_2 (UV/H_2O_2), the colour and COD removals can be achieved up to 98.77% and 86.11%, respectively, using 0.6 M of H_2O_2 at pH 6 for 81 min [84]. In this method, the use of UV rays helps to facilitate the decomposition of H_2O_2 and forms a high concentration of $OH^•$ radicals, which eventually increases the colour and COD removal from wastewater. Despite that, this method is limited to certain types of dyes and is sensitive to scavenging effects. As UV rays are applied to the process, the turbidity of the wastewater may be affected, leading to high operating and maintenance costs. As for the Fenton reaction, this method is known to be highly reactive in attacking dyes and provides a faster reaction compared to other oxidation processes. A study [85] revealed that mixed equal ratios of different reactive dyes at pH below 3.5 exhibit similar colour and COD removals by 97% and 90%, respectively. This indicates that the Fenton reaction is highly effective in reducing COD compared to other AOP. Nevertheless, the presence of ferrous ions within the process may lead to the generation of iron sludge. Also, this method may lead to operational problems as it requires suitable dosage ratios to effectively remove reactive dyes from wastewater.

All in all, AOP is favoured and is widely used due to its effectiveness in generating highly active oxidising agents for the removal of reactive dyes from wastewater. Despite that, these methods are limited to the formation of $OH^•$ radicals, and the decolourisation and degradation rate of dyes are greatly affected by the pH and dosage of oxidising agents used. Moreover, AOP is uneconomical as it results in high treatment costs due to the need to add chemical reagents and high energy consumption. Also, these methods may be complex and costly due to the sludge generation process, which may be hazardous, necessitating another treatment process before disposing of the sludge. Like conventional oxidation processes, these methods are dangerous to transport and store. This has made researchers shift to the application of hybrid or combined treatment processes to improve the disadvantages of these individual processes.

3.6. Electrochemical processes

Because advanced oxidation processes (AOP) are expensive and frequently cause operational problems, extensive research has been conducted in which the applications of electrochemical (EC) processes have piqued the interest of researchers due to their promising effectiveness in reducing colour, turbidity, dissolved solids and COD from wastewater without forming solid residues [86–88]. The advantages offered by EC processes, such as simplicity in operation and comparable treatment costs compared to other treatment methods, have also made it a point of interest and it is often used in industries. The applications of EC processes can be classified into a few different processes which include electrocoagulation, electrooxidation, and photo-electrochemical [34,89]. These methods are known to provide cleaner energy as the redox reactions that take place on the surface of electrodes are feasible in destroying organic compounds, almost completely converting them into carbon dioxide and water [34]. Among all the electrochemical processes, the electrocoagulation process is known to be the most established [90]. The applications of electrochemical processes to remove colour from wastewater are shown in Table 5.

In general, EC takes place in an electrochemical cell in which electric current is supplied between sacrificial metal electrodes (anode and cathode) that are immersed in an intermediate space filled with electrolyte. Within the reactor, oxidation and reduction processes take place at the anode and cathode, respectively [88,91]. Factors such as pH, reaction time, current density, conductivity, and selection of electrode materials greatly affect the performance and the treatment efficiency of EC processes [86]. Initially, the mechanism of EC processes involves the generation of metal ions on an anode surface. This eventually leads to the formation of coagulating agents such as monomeric and polymeric hydroxyl complexes. With the continuous supply of electric current, these metal complexes will initiate the coagulation process and form flocs that entrap pollutants to be removed from the process. Simultaneously, at the cathode, hydroxyl ions will be formed, releasing hydrogen gas bubbles that carry light solids and hydrophobic materials that will be removed from wastewater through sedimentation or floatation [10,86]. The reaction that occurs within the EC process can be expressed in Eqs. (7)–(9) whereby *M* represents the electrode types [86]. Aluminium and iron are commonly used as sacrificial electrodes because they are inexpensive, widely available and highly effective at removing dyes from wastewater [11,88,92].

At anode:

$$M_{(s)} \rightarrow M^{n+}_{(aq)} + ne^- \quad (7)$$

$$2H_2O \rightarrow 4H^+_{(aq)} + O_{2(g)} + 4e^- \quad (8)$$

At cathode:

$$nH_2O + ne^- \rightarrow \left(\frac{n}{2}\right)H_{2(g)} + nOH^-_{(aq)} \quad (9)$$

Over the years, many efforts have been made to optimise the removal of dyes from textile wastewater using EC processes such as electrocoagulation, electrooxidation, and photo electrochemical. Compared to conventional treatment methods, they are known to be more robust and compact, making them potentially effective in treating large volumes of wastewater. In reference to Table 5, a study from [3] has investigated the potential of the electrocoagulation process using iron and aluminium as electrodes for both anode and cathode for a reaction time of 3 h. It was revealed that both electrodes achieved similar colour and COD removals of 92% and 88%, respectively. Another study [87] also examined the performance of electrocoagulation using an aluminium electrode at pH 4.5 with a current density of 40 A/m^2 for 10 min of operating time. A comparison has been made with the study conducted by [3] in which this method was proven to not only be effective in removing colour and COD but also be able to remove TOC, total suspended solids (TSS), and turbidity from wastewater. On the contrary, most of the studies for the electrochemical oxidation method use platinum as an electrode and have proven to be promising in removing COD for different types of reactive dyes by up to 90% [3,34,93]. As for the photo-electrochemical method, it was revealed that colour and TOC were able to be removed by 99% and 11%, respectively [94]. These results prove that EC processes are effective in treating various reactive dyes from wastewater.

In short, the advantages of EC processes, such as easy operation, producing less sludge, and minimal use of chemicals, have made them attractive in the treatment of wastewater, as unfeasible capital costs can be prevented. The use of these methods in industries has also been proven highly reliable as they require less retention time and are capable of eliminating a wide range of organic and inorganic pollutants from wastewater compared to other treatment processes. Nevertheless, there is a need for electrode maintenance as the electrodes used may corrode over time. Moreover, these processes may consume large amounts of energy for operation, making them costly. Hence, further research has been carried out to combine the EC process with other treatment processes for better treatment performance.

3.7. Advantages and disadvantages of existing textile wastewater treatment processes

The accretion of reactive dyes in textile industries has always posed a grave problem in which, even at low concentration, reactive dyes will destroy the aesthetic value of water and, undoubtedly, deteriorate the environment as they are discharged into water bodies. As a result, leading researchers have devised a wide range of treatment methods, including biological, physico-chemical, electrochemical, conventional, and advanced oxidation processes, to ensure that industrial effluent can be safely discharged into the environment. Factors such as the nature or characteristics of the wastewater, treatment costs, and operational

Table 5
Applications of electrochemical processes in treating textile wastewater

Process	Dye	Operating conditions	% Removal	References
Electrocoagulation	Dye effluent	Using Fe–Fe electrode at 8 V for 3 h Using Al–Al electrode at 10 V for 3 h	Colour = 92, COD = 88 Colour = 92, COD = 88	[3]
	Blue reactive dye	Operated using an Al electrode at pH 10 with a current density of 10 mA/cm^2 for an electrolysis time of 60–120 min	Colour = 90 to 95 COD = 30 to 36	[4]
	Basic Red 18	Operated using an Al electrode at pH 7, 50 V, with a reaction time of 60 min, a conductivity of 3,000 µS/cm and an initial dye conc. of 50 mg/L	Colour = 97.7	[35]
	Reactive Yellow 135	Operate using an Al electrode at pH 4.5, a current density of 40 A/m^2 and a 10 min operating time	COD = 81 TOC = 85 TSS = 97.1 Turbidity = 93.7	[87]
Electrochemical oxidation	Dye effluent	Using Pt–Fe electrode at 6 V for 3 h	COD = 93	[3]
	Reactive Blue 52 Reactive Black 5 Reactive Green 15 Reactive Yellow 125	Operated using a Pt electrode and an initial conc. of 200 mg/L at 12 V for an electrolysis time of 60 min	All COD values were reduced by up to 30 mg/L O_2, except RB5	[34]
	Reactive Orange 16	Operated using a Pt electrode and the removal efficiency of dye was tested in acid and basic media for 4 h	Removal of colour in acid and basic media is 40% and 18%, respectively	[93]
Photo-electrochemical	Reactive dyes	Operated using 10 A of current density for 10 min	Colour = 99 TOC = 4 to 11	[94]

feasibility are considered in utilising the performance of a treatment process. However, compared to other dye classes, reactive dyes are known to be difficult to degrade, thus making it challenging for industries to choose the appropriate treatment to obtain satisfactory removal of reactive dyes. In this section, the advantages and disadvantages of the treatment processes for the removal of dyes from wastewater will be discussed.

Table 6 presents the summary of advantages and disadvantages of existing treatment processes in textile wastewater treatment. The use of biological treatment processes such as aerobic and anaerobic processes is highly attractive in the treatment of textile wastewater due to the presence of extremophilic microorganisms used within the process that can remove dyes from wastewater. However, with the increasing number of textile industries, this method becomes uneconomically feasible as large spaces are required for installation. Also, the slow reaction time and the high polymeric molecular structure of reactive dyes that are toxic and resistant to biological organisms make it difficult to degrade reactive dyes through biological treatment processes. The public's growing concern about the limitations of biological approaches, which could lead irresponsible parties to discharge untreated textile effluent into water bodies, has shifted to the use of physico-chemical processes.

For physico-chemical processes such as the coagulation process, it may easily decolourise insoluble dyes like dispersed dyes but is inefficient in treating water-soluble dyes like reactive dyes. Also, the addition of chemicals within the process may generate a large amount of sludge and produce toxic by-products, hence making it expensive as post-treatment processes may be required to overcome disposal problems. Another popular wastewater treatment process is adsorption, which is highly effective in removing various types of dyes. Despite that, adsorption is a complicated and expensive process as pre-treatment of the adsorbent is required before inserting them into the adsorption column. Also, the process may require high maintenance for adsorbent regeneration and spent adsorbent disposal. Like the adsorption process, membrane filtration is also commonly used in treating textile wastewater due to its simplicity for maintenance, ability to remove various types of dyes, no sludge generation, and less space for operation. However, fouling might occur in membrane technology, which eventually decreases the removal efficiency over time and may also lead to high operational and maintenance costs. Additionally, the applications of membranes that are bound to treat small volumes of textile wastewater also limit their application on a larger scale.

It can be concluded that the environmental impacts and the complex structure of reactive dyes that are resistant to biological organisms and chemicals in nature make conventional treatment processes such as biological and physico-chemical processes inefficient. Alternatively, chemical oxidation processes involving the use of conventional and advanced oxidation processes were introduced. Chemical oxidation processes involve the use of oxidising agents such as O_3 and H_2O_2 that help in generating $OH^•$ radicals that help in degrading organic compounds into carbon dioxide, mineral acids, and water. However, these chemical oxidation processes are uneconomical as the use of oxidising agents may result in high treatment costs and lead to operational and environmental problems. The limitations faced using chemical oxidation processes are later overcome using electrochemical processes.

Electrochemical (EC) processes such as electrocoagulation have been actively emerging due to their versatility in eliminating broad ranges of organic pollutants from textile wastewater. Compared to chemical oxidation processes, EC is more effective and cheaper as it requires minimal use of chemicals. Despite that, the applications of EC processes are limited as the use of sacrificial anodes like iron may only transfer pollutants at the bottom of the reactor instead of degrading them to be removed completely from the process. Also, the oxidation efficiency in EC processes is relatively low, and the high energy consumption may lead to high operational costs. All these factors indicate that, unfortunately, there are no treatment processes that can universally remove dyes from textile wastewater.

Recently, extensive research has been carried out by researchers in search of an appropriate textile wastewater treatment by considering the merge of individual treatment processes containing biological, physical, and chemical processes. The potential of hybrid or combined treatment processes as an interesting option in reducing toxic or inhibitory bacterial activities and removing dyes from wastewater has made it a point of interest. The applications of hybrid or combined treatment processes in the decolourisation of dyes will be further discussed in the next section.

3.8. Hybrid or combined processes

At present, existing individual treatment processes such as biological, physico-chemical, electrochemical, conventional, and advanced oxidation processes alone are inadequate for removing reactive dyes from textile wastewater [53]. This is due to the nature or characteristics of reactive dyes that are complex and difficult to degrade. Also, textile wastewater may contain a high concentration of COD (approximately 50–18,000 mg/L), thus requiring additional techniques [100]. Conventional methods such as biological and physico-chemical processes are only involved in transferring organic pollutants from one phase to another instead of removing them completely from the process. This consequently leads to the production of toxic by-products, resulting in need for further treatment of sludge disposal or the wastewater itself. Meanwhile, the use of oxidising agents in chemical oxidation processes may result in high treatment costs and lead to operational and environmental problems. On the other hand, the applications of electrochemical processes are limited due to the low oxidation efficiency and high energy consumption, which later lead to high operational costs.

Hence, extensive research has been carried out in search of more effective and cheaper treatment processes for the treatment of textile wastewater. Recently, the practise of using hybrid or combined treatment processes has raised interest among researchers due to its potential to minimise the disadvantages of individual processes and further improve the rate of dye removal from wastewater. Over the years, many efforts have been made by researchers to achieve the discharge standards by introducing various hybrid processes, including the combination of physical/chemical-biological and physical/chemical-advanced oxidation processes. Table 7 reviews the research that has been attempted in the application of hybrid processes for textile wastewater treatment.

As shown in Table 7, recently, many efforts have been made in the development of hybrid or combined treatment processes for the removal of dyes from textile wastewater. In general, hybrid or combined treatment processes refer to the combination of two or more individual processes that have the potential to minimise the disadvantages of individual processes and further improve the effectiveness of the overall treatment process [104]. Compared to conventional treatment processes, they are known to be more effective in removing various types of pollutants from wastewater. A study [101] investigated the potential of combining adsorption and coagulation processes and revealed that colour can be removed up to 96.67% using *Hibiscus sabdariffa* seeds as coagulant and activated carbon at pH 2 with a dosage of 209 mg/L. Another study [24] proposed the process of combining adsorption and ultrafiltration processes. It is reported that colour and COD can be removed by 97% and 70%, respectively, along with permeate flux of 450 L/h m^2 at pH of 7, transmembrane pressure of 3 bar and 150 mg/L dosage of PAC in the presence of cationic surfactant at 25°C for 180 min. These studies show that employing the combination of two different physico-chemical processes is effective in achieving maximum adsorption capacity.

The study on combining two different electrochemical processes was also developed by integrating electrocoagulation and electrooxidation processes [102]. In this study, it was found that complete removal of colour can be achieved and pollutants such as DCOD and TOC are able to be removed by operating the hybrid process in a multistage reactor using an aluminium electrode at pH 4, conductivity of 3.7 mS/cm and a current density of 4.1 mA/cm^2 for 90 min. An attempt to combine electrocoagulation with nanofiltration was also carried out by [100]. The usage of these combinations is also effective in removing colour and turbidity by 95% and 80%, respectively, with a retention rate of more than 92% using an aluminium electrode with a current density of 40 mA/cm^2, an electrolysis time of 60 min, and a transmembrane pressure of 10 bar at a temperature of 30°C. [103] also investigated the efficacy of peroxi- and ultrasonic electrocoagulation in the treatment of textile wastewater. Similar to the study conducted by [102], this hybrid process is able to remove colour, COD and TOC from wastewater using an iron electrode at pH 3 with a US frequency and power of 37 kHz and 256 W, respectively. As for the combination of ultrafiltration with electrodialysis, the process may only remove total dissolved solids (TDS) by 94.2% at pH 5.3 with a conductivity of 0.45 mS/cm, a flow rate of 10 L/h and a voltage of 10 V for 25 min. These results indicate that these hybrid processes are effective in treating various reactive dyes from wastewater.

Table 6
The summary of advantages and disadvantages of existing textile wastewater treatment processes

Type of treatment process	Advantages	Disadvantages	References
		Biological treatment processes	
Aerobic process Anaerobic process	Is efficient for the removal of azo dyes Low sludge production, and sludge can be recycled High BOD and COD removal	Insufficient in treating textile wastewater Slow reaction time, depending on the type of strain or culture High operating and maintenance costs It requires large spaces	[25], [30], [33], [95], [96]
		Physico-chemical processes	
Coagulation–flocculation	Fast reaction Simple operation High removal of BOD and COD efficiency	High sludge production It requires high costs for chemical usage This causes handling and disposal problems	[25], [30], [33], [60]
Adsorption	Applies to various types of dyes or industries Simple operation High BOD and COD reduction	The efficiency of the adsorbent is lost over time Cause utilised adsorbents disposal problems Regeneration of the adsorbent is expensive	[33], [60], [95], [97]
Membrane filtration	Minimal use of chemicals High BOD and COD removal with a multistage system It is able to decolourise most types of dyes	A large amount of sludge is produced High operating and maintenance costs Fouling occurs and removal efficiency decreases over time	[25], [95], [97]
		Electrochemical processes	
Electrocoagulation Electrooxidation	More effective than the conventional coagulation method Minimal use of chemicals Low sludge production High removal of colour Short retention time Simple operation	High operating and maintenance costs Electrodes may need to be changed on a regular basis.	[30], [31], [58], [60], [95], [97], [98]

(Continued)

Table 6 Continued

Type of treatment process	Advantages	Disadvantages	References
	Conventional oxidation processes		
Ozonation	There is a reduction in the production of hazardous waste	High operating and maintenance costs	[30], [31], [33], [50], [58], [60]
Hydrogen peroxide	No sludge production	Low COD removal	
		Short half-life and is not suitable for dispersed dyes	
	Advanced oxidation processes		
Ozone-based			
H_2O_2/O_3	No production of sludge	Expensive	[99]
	The non-selective hydroxyl radicals formed are strong enough to break down conjugated double bonds	May lead to the formation of toxic by-products	
		Few hydroxyl radicals are formed	
O_3/UV	Higher efficiency compared to O_3 or UV processes alone	It requires high energy and cost	[99]
	It generates more hydroxyl radicals than H_2O_2/UV	The use of UV light may be obstructed by the turbidity of the wastewater	
	Hydrogen peroxide-based		
H_2O_2/UV	No potential formation of bromated compounds	The use of UV light may be obstructed by the turbidity of the wastewater	[99]
	May be used fully as a treatment system for drinking water	The presence of compounds such as nitrate may affect the UV light absorbance	
Fenton reaction	Effective for the removal of both soluble and insoluble dyes	Leads to sludge generation	[31], [50], [58], [60], [98], [99]
	High colour removal efficiency	High chemical and energy consumption	
		High operating costs as an increase in pH adjustment	

Table 7
Applications of hybrid or combination processes in treating textile wastewater

Type of hybrid process	Dye	Operating conditions	% Removal	References
Adsorption/coagulation	Congo Red	*Hibiscus sabdariffa* seeds are used as coagulant and activated carbon at pH 2 with a dosage of 209 mg/L	Colour = 96.67	[101]
Adsorption/ultrafiltration	Acid Orange 7	At 25°C for 180 min, the pH was set to 7, the transmembrane pressure was set to 3 bar, and the PAC dosage was 150 mg/L in the presence of cationic surfactant	Permeate flux = 450 L/h m^2 Colour = 97 COD = 70	[24]
Electrocoagulation/ electrooxidation	Denim effluent	Aluminium electrode with pH 4, conductivity 3.7 of mS/cm and current density of 4.1 mA/cm^2 for 90 min in a multistage reactor	Colour = 100 DCOD > 70 TOC > 60 BOD_5/COD = 0.44	[102]
Electrocoagulation/ nanofiltration	Dye effluent	Operated using an aluminium electrode with a current density of 40 mA/cm^2, an electrolysis time of 60 min, and a transmembrane pressure of 10 bar at a temperature 30°C	Colour = 90 to 95 Turbidity = 80 Retention rate > 92	[100]
Peroxi- and ultrasonic electrocoagulation	Direct Red 31	Iron electrode is used at pH 3 with US frequency and power of 37 kHz and 256 W, respectively	Colour = 93.3 COD = 86.7 TOC = 58.7	[103]
Ultrafiltration/ electrodialysis	Dye effluent	Operate at pH 5.3 and conductivity of 0.45 mS/cm, flow rate 10 L/h and voltage of 10 V for 25 min	TDS = 94.2	[38]

In short, compared to the individual processes, high removal of colour, COD, TOC, and TDS percentages can be achieved using the hybrid or combined treatment processes. Also, these hybrid processes are attractive as they are easier to regulate or maintain. The reaction is also faster with minimal usage of chemicals and less energy consumption. The advantages offered by these hybrid processes have proven that they are effective in treating textile wastewater. Despite that, in Malaysia, till date, the research effort sequencing the use of hybrid or combined treatment processes is still lacking. Upon review, it is believed that research on the use of this technology in the treatment of textile wastewater will continue to be further explored in the future.

4. Conclusion

Overall, it is clear that the goals of developing cheaper, more effective, and novel treatment processes have pushed researchers to become more involved in textile wastewater treatment. In this work, different types of treatment processes along with their advantages and disadvantages for the removal of reactive dyes from textile wastewater have been exposed. Reactive dyes have always been known to be difficult to degrade, thus making it challenging for industries to choose the appropriate treatment to obtain satisfactory removal of reactive dyes. Due to economic factors and in meeting the discharge standards, the use of existing treatment processes such as biological and physico-chemical processes alone have gained the attention of most textile industries in Malaysia. However, the complex and recalcitrant structures of reactive dyes have led to the incomplete destruction of organic matters and the limitations of these treatment processes, which include slow reaction time, the nature of reactive dyes that are resistant towards biological organisms and require a certain amount of chemicals, make them insufficient in removing reactive dyes from wastewater. Also, these conventional treatment processes may only involve the transfer of organic pollutants from one phase to another instead of removing them completely from the process. This later leads to the generation of toxic by-products, causing many industries to face problems in abiding by environmental standards. The need for further treatment for sludge disposal and discharge of the wastewater itself makes these treatment processes expensive. With the increasing number of textile industries, the limitations of these conventional treatment processes may result in irresponsible parties discharging the untreated textile effluent into water bodies. This consequently leads to extensive research in search of more effective and cheaper treatment processes for the treatment of textile wastewater.

Alternatively, the trend of textile wastewater treatment then continues to develop into the use of chemical oxidation processes involving the use of conventional and advanced oxidation processes. In these processes, oxidising agents such as O_3 and H_2O_2 are used to generate $OH^{\bullet}$ radicals, which later help in degrading organic compounds into carbon dioxide, mineral acids, and water. However, these chemical oxidation processes later create another problem as they may result in high treatment costs and lead to operational and environmental problems. The limitations faced using chemical oxidation processes are later overcome using electrochemical (EC) processes. The use of EC processes such as electrocoagulation has been actively emerging due to its versatility in eliminating broad ranges of organic pollutants from textile wastewater. EC is known to be more effective and cheaper compared to chemical oxidation processes as it requires minimal use of chemicals. Despite that, the applications of EC processes are limited as the use of sacrificial anodes like iron may only transfer pollutants at the bottom of the reactor instead of degrading them to be removed completely from the process. Also, EC processes have relatively low oxidation efficiency, and their high energy consumption may lead to high operational costs. Indirectly, all these factors indicate that, unfortunately, there are no treatment processes that can universally remove dyes from textile wastewater.

The continual rise of environmental problems due to the high concentration of COD in textile wastewater and the inefficiency of the existing treatment processes alone has led researchers to consider merging individual treatment processes containing biological, physical, and chemical processes. It has been underlined that the use of hybrid or combined treatment processes is a promising option in the treatment of textile wastewater due to its potential to minimise the disadvantages of individual processes and further improve the rate of dye removal from wastewater. Compared to existing wastewater treatment processes, the applications of hybrid or combined treatment processes are known to be effective in reducing toxic or inhibitory bacterial activities and removing dyes from wastewater. Based on extensive research that has been carried out, it is concluded that better treatment efficiency can be achieved by combining individual treatment processes. For instance, combining membrane technology with other treatment processes such as coagulation–flocculation, adsorption, and EC processes may reduce fouling, operating costs, and enhance the removal of dyes from wastewater. However, in Malaysia, the full-scale applications of hybrid processes are still in their infancy. Nevertheless, with the continuous research efforts and potential investment opportunities in the treatment of textile wastewater in Malaysia, it is expected that the use of hybrid or combined treatment processes in Malaysia will continue to grow rapidly and will be more significant in the future, given the effectiveness and removal efficiency obtained in past research.

Acknowledgement

The authors gratefully acknowledge the final support received from Universiti Teknologi MARA for the KEPU research funding of 600-RMC/KEPU 5/3 (011/2021), the Institute of Research Management and Innovation (IRMI), and the School of Chemical Engineering, College of Engineering, Universiti Teknologi MARA, Shah Alam, Selangor, Malaysia, in making this review viable and effective.

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