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A review of the textile wastewater treatment technologies with special focus on advanced oxidation processes (AOPs), membrane separation and integrated AOP-membrane processes

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ABSTRACT

The current work presents a review of some recent studies focussing on the treatment of textile wastewater using advanced oxidation processes (AOPs), membrane separation and the integrated AOP-membrane process. The textile industry is the most water-intensive industry and discharges a wide variety of pollutants with widely varying values into its effluents, which among other factors, depend on the wet processes undertaken at certain textile industry, the geographical location of the industry, the substrates involved, and the processing conditions used. The application of homogeneous and heterogeneous AOPs for treating textile wastewater has been reviewed. Unlike homogeneous AOPs, heterogeneous AOPs use transition metal-based catalysts, produce higher kinetics in shorter treatment times, and generate a lesser amount of sludge. However, the techno-economic optimization of these catalysts in both the homogeneous and heterogeneous AOPs is a key direction for further research work. On the other hand, membranes have shown better performances than the conventional treatment processes, produce lesser byproducts, and have been found to be a suitable integration option to have a more robust treatment process employing both the AOPs and membrane separations. However, more work is needed to enhance the throughput, anti-fouling properties and large-scale module design of these membranes. Based upon a handful of studies focussing on the use of integrated AOP-membrane separation processes to treat wastewater, various process configurations were proposed for future engineering design. Key challenges hindering the development of such integrated process concepts have been presented, which could work as a reference for future research and development.

Keywords: Textile; Wastewater; Membrane; Advanced oxidation processes (AOPs); Integrated process; Treatment; Review

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1. Introduction

With a growing global population and rapid socio-economic growth, clean water is becoming a scarce resource, thus highlighting the need and importance of water treatment to tackle this issue [1]. According to the Food and Agriculture Organization of the United Nations, the industrial sector consumed around 51% of the total water supply, followed by agriculture (36%) and municipal (13%) in the USA in 2010 [2,3]. The textile industry is considered a highly water-intensive industry and consumes around 2.1% of the industrial water, which corresponds to a freshwater consumption of as high as 0.40 m^3 per kg of the final product [4]. It is one of the highly polluting industries and the generated wastewater becomes a major environmental obstacle for the development of the textile industry as the textile industry uses many chemicals and discharges large amounts of highly colored wastewaters with high organic contamination and poor biodegradability, which make the treatment very difficult.

There are a number of studies focusing on various individual aspects of wastewater and wastewater treatment. However, there are only a handful of studies discussing two or more individual treatment technologies in detail, and then providing up-to-date insight into the use of their combination for tertiary treatment of wastewater [5]. Kiran et al. [6] provided a brief introduction of various technologies used to treat textile wastewater, and compared their advantages and disadvantages. Garcia-Segura et al. [7] studied the application of the Fenton process to wastewater treatment and detailed the kinetics and mechanisms involved in removing various pollutants. Ziajahromi et al. [8] studied the existence of microplastics in wastewater effluents and detailed the mechanism and effects of the interaction of microplastics with wastewater contaminants on aquatic life. Särkkä et al. [9] reviewed the application of electrochemical oxidation (EO) in wastewater treatment and emphasized the need of combining EO with other treatment technologies to obtain better water purification performance. Paździor et al. [10] reviewed the combination of biological treatment with advanced oxidation processes (AOPs) and reported that the overall performance of water treatment improved due to the combined process. They also reported case studies from the textile wastewater industry in Poland. The present work aims at providing information that can help to understand various pollutants present in the effluents emitted from various textile industry's processing steps with a focus on recent advances in applying AOPs, membrane technology and particularly their combinations to tertiary-treat textile wastewater. The work summarizes these combined membrane-AOP strategies and provides direction for future research work in this area.

1.1. Processing steps and corresponding pollutants in textile wastewater

The textile industry uses more water than any other industry in the world, which is required in its different wet processing operations. Fig. 1 shows the various processing steps involved in the wet processing of fabrics and their corresponding water consumption. Among these, bleaching and finishing consume the highest amount of water.

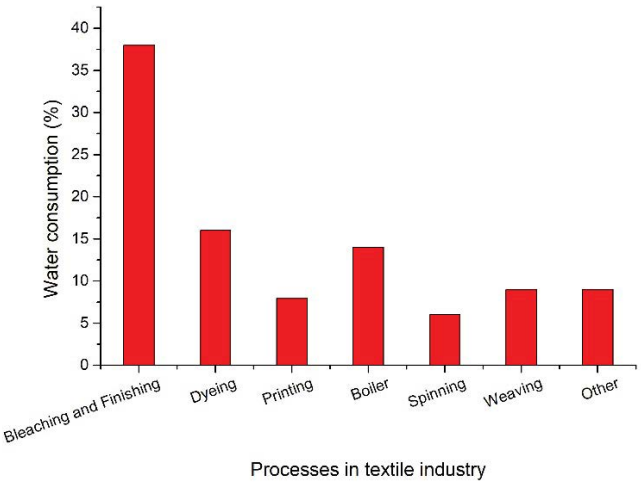

Fig. 1. Water consumption in various steps involving wet processing of fabric [11].

With regard to its impact on the ecosystem, it is reported that most of the discharged textile effluents are heavily polluted with various chemicals, such as acids, bases, dyes, surfactants, dispersing agents and metal ions. It is worth mentioning that the wastewaters produced from various processing steps are significantly different from each other, and textile wastewater often refers to mixed wastewater coming out from various processing steps of a textile factory.

Table 1 presents the main processing steps, substrates used, and the corresponding inorganic and organic pollutants discharged in the textile industry [12–14].

Sizing and desizing produce small volumes of highly polluted and concentrated wastewater. Generally, the chemicals used in these process steps include starch, polyvinyl alcohol (PVA), polyacrylates, carboxymethyl cellulose, alkalis, acids, enzymes or surfactants, and render the generated wastewater to be high in chemical oxygen demand (COD), biological oxygen demand (BOD) and suspended solids [15–17]. Scouring is used to remove impurities from fibers, and involves using scouring agents, such as detergents, soaps, alkalis, wetting agents, defoamers and lubricants, which result in significant contributions to COD and suspended solids in textile wastewater [18]. Carbonising involves treating the fibers with acid to remove greases and degrade cellulose at high temperatures, which result in wastewater that is low in organic content, but high in dissolved solids [19]. After carbonizing, the fabric is treated with soda ash or sulphuric acid to make it denser (felting), which results in high BOD levels in the generated wastewater [20]. Bleaching is conducted to remove natural colors from fabric and includes the use of sodium hypochlorite and hydrogen peroxide, which contribute to high suspended solids' loading in the wastewater. Dyeing and printing are two of the most important process steps in the textile industry and impart color, metals and salts to the generated wastewater [21].

It is obvious that the textile industry discharges wastewater with a wide range of pollutants. Many organic pollutants are biodegradable, however many others are recalcitrant due to their complex chemical structure and

Table 1
Various processing steps of the textile industry, substrates used, and inorganic and organic pollutants in the corresponding effluent [12–14]

Process step	Substrates used	Inorganic pollutants		Organic pollutants (biodegradability)
Desizing	Cotton	Na^+	SO_4^{2-}	Carboxymethyl cellulose (SB); enzymes (A); fats (SB); hemicelluloses (A);
	Linen	Ca^{2+}	Cl^-	modified starches (B); non-ionic surfactants (A); oils (SB); starch (B); waxes (SB)
	Viscose	NH^{4+}		
	Silk	Na^+	CO_3^{2-}	Carboxymethyl cellulose (SB); enzymes (A); fats (SB); gelatine (A); oils (SB);
	Acetates	NH^{4+}	PO_4^{3-}	polymeric sizes (NB); polyvinyl alcohol (A); starch (B); waxes (SB)
	Synthetics			
Scouring	Cotton	Na^+	CO_3^{2-}, PO_4^{3-}	Anionic surfactants (A); cotton waxes (NB); fats (SB); glycerol (B); hemicelluloses (A); non-ionic surfactants (A); peptic matter (A); sizes (A); soaps (A); starch (A)
	Viscose acetates	Na^+	CO_3^{2-}, PO_4^{3-}	Anionic detergents (B); fats (SB); non-ionic detergents (B); oils (SB); sizes (B); soaps (B); waxes (SB)
	Synthetics	Na^+	CO_3^{2-}, PO_4^{3-}	Anionic surfactants (A); anti-static agents (NB); fats (SB); non-ionic surfactants (A); oils (SB); petroleum spirit (A); sizes (B); soaps (A); waxes (SB)
	Wool (yarn and fabric)	Na^+, NH^{4+}	CO_3^{2-}, PO_4^{3-}	Anionic detergents (A); glycol (SB); mineral oils (SB); non-ionic detergents (A); soaps (A)
	Wool (loose fibre)	Na^+, NH^{4+}, K^+, Ca^{2+}	CO_3^{2-}, PO_4^{3-}, Cl^-	Acetate (B); anionic surfactants (A); formate (B); nitrogenous matter (U); soaps (A); suint (A); wool grease (SB); wool wax (SB)
Bleaching	Cotton	Na^+	ClO^-	Formate (B)
	Linen	NH^{4+}	Cl^-	
	Viscose		O_2^{2-}	
	Jute		F^-, SiO_3^{2-}	
	Synthetics		SiO_3^{2-}	
	Acetates		PO_4^{3-}, F^-	
	Wool	Na^+	O_2^{2-}	Oxalate (B)
Mercerizing	Cotton	Na^+	CO_3^{2-}	
	Linen	NH^{4+}	SO_4^{2-}	

(Continued)

Table 1 Continued

Process step	Substrates used	Inorganic pollutants		Organic pollutants (biodegradability)
Dyeing	Cotton Viscose Linen	Na^+, Cr^{3+}, Cu^{2+}, Sb^{3+}, K^+, NH^{4+}	Cl^-, CO_3^{2-}, CO_4^{2-}, F^-, NO^{2-}, O_2^{2-}, S^{2-}, $S_2O_3^{2-}$, SO_3^{2-}, SO_4^{2-}	Naphthol (A); acetate (B); amides of naphthalic acid (B); anionic dispersing agents (A); anionic surfactants (A); cationic fixing agents (NB); chloro amines (SB); formaldehyde (A); formate (B); nitro amines (SB); non-ionic surfactants; residual dyes (NB); soaps (A); soluble oils (SB); sulphated oils (A); tannic acid (A); tartrate (B); urea (B)
	Wool	Na^+, K^+, NH^{4+}, Cr^{3+}, Cu^{2+}, Al^{3+}, Sb^{3+}	SO_4^{2-}, SO_3^{2-}, S_2O^{4-}, CO_3^{2-}, Cl^-	Acetate (B); dispersing agents (U); formate (B); lactate (B); residual dyes (NB); sulphonated oils (A); tartrate (B)
	Polyamide	Na^+	Cl^-, CO_3^{2-}	Acetate (B); formate (B); polyamide oligeines (NK); residual dyes (NB); sulphonated oils (A)
	Acrylic	Na^+, Cu^{2+}, NH^{4+}	SO_4^{2-}	Acetate (B); aromatic amines (A); formate (B); levelling agents (NK); phenolic compounds (A); residual dyes (NB); retardants (NK); surfactants (A); thioreia dioxide (A)
	Polyester	Na^+, NH^{4+}	$S_4O_6^{2-}$, ClO^-, NO^{3-}, Cl^-, SO_3^{2-}	Acetate (B); anionic surfactants (A); anti-static agents (NB); dispersing agents (A); dye carriers (SB); ethylenediaminetetraacetic acid (NB); ethylene oxide condensates (NK); formate (B); mineral oils (SB); non-ionic surfactants (A); residual dyes (NB); soaps (A); solvents (A)
Printing and finishing	Cotton, wool, polyester, acrylic, nylon 6	NH^{4+}	SO_4^{2-}, SO_3^{2-}	Polysaccharides B); carboxymethyl cellulose (CMC) derivatives (B); polyacrylates (SB); glycerine and polyols (B); polyvinyl alcohol (B); aromatics (NK); mineral oils/aliphatic hydrocarbons (NK)

B stands for biodegradable; A for biodegradable after acclimatization; NK for not known; NB for non-biodegradable; SB for slowly degradable.

synthetic organic origin, thus making the treatment process very challenging.

1.2. Characteristics of textile wastewater and various discharge standards

The quality of effluent is characterized based upon certain parameters, mainly including color, pH, electrical conductivity (EC), COD, BOD and total dissolved solids (TDS) [22]. Various other parameters, such as contents of oil and grease, chloride, phenol, fluoride, phosphate, silica, sodium and certain heavy metals including Cu, Pb, Mn and Cd are also included in some studies [23,24]. The values of these parameters vary based upon the process concept employed in various textile mills, time of the sampling, type of the product produced, type of the chemicals used, and the geographical location of the industry [25]. Tables 2a and 2b present the characteristics of various wastewaters originating from different factories located in different geographical territories of the world.

In order to give more meaning to data presented in Tables 2a and 2b, it is worthwhile to know various discharge standards that are used in various parts of the world. Table 3 presents the permissible limits for various textile wastewater effluents to be discharged in the ecosystem.

Based upon the most-relaxed permissible limits presented in Table 3, the values given in Tables 2a and 2b can help understand the nature of pollution in textile wastewater. It could be seen that in 9 (out of 23) studies, the pH values of the wastewater lied beyond the permissible range (5–9). This could be due to the type and strength of acid and alkali dyes used during the dyeing process. Parameters of BOD, COD and total suspended solids (TSS) are generally higher than the permissible limits, which could be due to the chemicals used during sizing/desizing operations. Out of 12 studies, 9 reported higher TSS values than the permissible limits given in Table 3. Sodium is reported to be very high (four out of four studies reporting higher values) as compared to the permissible limit, as shown by the data presented in Tables 2a and 2b. However, the concentrations of heavy metals, such as Zn, Cr, Cu, and Fe seemed to be within the permissible limits (data not shown). This is particularly evident for Cu (four out of four data sets falling within the permissible limits), Cr (two out of three datasets falling within the permissible limits) and sulfate (six out of seven datasets falling within the permissible limits). Although this analysis is based upon randomly selected studies (Tables 2a and 2b) and portrays a grave picture of textile wastewater contamination, it should be emphasized that the discharge standards depend on a number of factors and vary considerably across various geographical and process industries.

It is generally accepted that if the ratio of BOD/COD is equal to or greater than 0.4, the wastewater can be treated using biological methods, whereas a value of 0.2–0.4 is considered to be representing partially degradable wastewater [54,55]. However, the review in current work (Tables 2a and 2b) shows that only three out of nine effluents have a BOD/COD ratio of more than 0.4, whereas six effluents lie significantly below this threshold (values ranging from 0.1–0.27). Such low values of the BOD/COD ratio indicate that these effluents cannot be treated using stand-alone biological technologies, and a supplementary method or technology needs to be employed to aid the biological treatment of such effluents. AOPs have shown favorable characteristics for treating non-biodegradable wastewaters, particularly the water reuse when combined with membrane technologies. The preceding sections further elaborate on the combination of AOPs and membrane technologies for treating textile wastewater.

2. Strategies for treating textile wastewater

Textile wastewater is high in color, low in BOD/COD ratio and high in salts (TSS and TDS). The situation is worsened by the fact that most textile effluents (particularly the ones coming from cotton, silk, and wool industries) contain reactive dyes, which are hard to biodegrade. These characteristics mean that different approaches have to be used to efficiently remove these pollutants. Fig. 2 presents the most common techniques used in treating textile wastewaters.

Colour is one of the most important parameters for wastewater effluents and restricts the penetration of sunlight into a water body, thus inhibiting photosynthesis and growth of aquatic species. It is well known that color is generally associated with the organic contaminants present in the effluent. In order to remove color and COD, and treat textile wastewater, various physical, biological and oxidative methods have been explored.

Coagulation–flocculation uses the addition of chemicals to alter the state of dissolved and suspended particulate matter in wastewater and separates them through sedimentation [56]. Various coagulants used to treat textile wastewater include hydrolyzing metallic salts (ferric chloride [57], ferric sulfate [58], magnesium chloride [59] and alum [60]), pre-hydrolyzing metallic salts (polyaluminium chloride [61], polyferric chloride [62], and polyaluminium sulfate [63]) and synthetic cationic polymers (aminomethyl polyacrylamide [64], polyalkylene [56], polyamine [65] and polyethyleneimine [66]). The method has been extensively used as a conventional pretreatment step for textile wastewater. However, the method suffers from the drawbacks of not being able to treat reactive dyes, while it also produces large volumes of sludge.

Adsorption has shown better results with regards to treating dye-containing wastewater, which is due to the higher affinity of dyes towards various adsorbents and the ability of the process to degenerate and reuse adsorbents in a continuous mode [67]. Various adsorbents, such as activated carbon [68], peat [69], bentonite clay [70], fly ash [47], and polymeric resins [71] have been used to treat textile wastewaters. Similar to coagulation–flocculation method, adsorbents suffer from the drawback of safe disposal, high cost and possible secondary pollution associated with the adsorbent regeneration.

Biological methods have extensively been used to treat dissolved matter present in textile wastewater and are generally applied when the BOD/COD ratio is equal to or higher than 0.4. These methods include aerobic, anaerobic or a combination of both [72]. A combination of anaerobic and aerobic biological treatment is the most common approach to treat textile wastewater, where the former lowers the high COD of wastewater, which is further reduced through aerobic treatment. Anaerobic treatment holds the advantage

Table 2a
Characteristics of real textile wastewater originating from different geographical locations

Source	Algeria [26]	Greece [27]	Mexico [28]	Thailand [29]	Brazil [30–32]			Turkey [33]		Iran [34]	Spain [35,36]	
Temperature (°C)									18–22			
pH	7.86	8.8	8.3	8.32–9.50	8.29	9	7.11	6.89	6.5–7.1	6.50	7.32	7.11
Colour (Pt-Co)	>200		10,000									
COD (mg/L)	340	470	975	278–736	550	482	729	305	1,940–2,060	1,310	2,100	708
BOD (mg/L)	210[a]		98	137[a]						180[a]		
EC (μS/cm)	3,120	7,200	5,700		725	1,410	273		2,250–2,350	1,260		3,840
TS (mg/L)				1,800–6,460	887		558					
TSS (mg/L)	300	68	130	85–354					210–250		52.05	
TDS (mg/L)				1,715–6,106								3,137
TOC (mg/L)		120						76	520–450		465	
Turbidity (NTU)	130						172				75.75	4.02
Volatile solids (mg/L)			100		489		378					
N–NH_3 (mg/L)			40.6									
Chloride (mg/L)						250						>125
Sulfate (mg/L)												28
Nitrite (mg/L)												
Nitrate (mg/L)												
Sodium (mg/L)												1,008
Magnesium (mg/L)												18.6
Potassium (mg/L)												
Phosphate (mg/L)												

EC, TS, TSS, TDS and TOC stand for electrical conductivity, total solids, total suspended solids, total dissolved solids and total organic carbon, respectively.
[a]specifically stated 5-d based BOD.

Table 2b
Characteristics of real textile wastewater originating from different geographical locations

Source	Malaysia [37]	India [38–41]				Taiwan [42]	China [43]	Portugal [44,45]			Ecuador [46]	Romania [47]
Temperature (°C)	35–58	38–40						31.4	30.9	20.5		
pH	3.85–11.40	8–10	10.2	10	9.2	10.8	8.5–9.0	11.3	8.1	11.1		7.30
Colour (Pt-Co)	169.67–1,937.33		962	1,410	610	868					80,800	389.86
COD (mg/L)	231.67–990.00	1,400–1,600	1,840	1,156	1,845		108–880	1,200	496	1,257	45,600	665
BOD (mg/L)			1,630	196		128		200[a]		200		386[a]
EC (μS/cm)	690–13,810		3,800		1,220		11,600–19,880	18,000	13,600	19,200		
TS (mg/L)												
TSS (mg/L)	22.67–150.00	180–200	496	8,430				<3	101			289
TDS (mg/L)	14.00–11,564.00	3,700–3,900	3,478	9,640	720			900				
TOC (mg/L)									129	390		
Turbidity (NTU)			209									93
Volatile solids (mg/L)	54.46–531.00								78			
N–NH_3 (mg/L)	0.47–50.83							32.5	8.8	32.5		
Chloride (mg/L)					325		1,122–1,138	5,500	1,100	5,100		98
Sulphate (mg/L)					124		1,568–3,405	119	400	160		365
Nitrite (mg/L)								<0.2	4.1	<0.2		
Nitrate (mg/L)	1.23–5.60						255–1,137		<0.2	3.4	3,610	
Sodium (mg/L)								4,100	1,200	4,200		
Magnesium (mg/L)								6	8.7			
Potassium (mg/L)								180	30.1			
Phosphate (mg/L)	0.07–4.01								6.4		558	

[a]specifically stated 5-d BOD.

Table 3
Permissible limits for various textile wastewater pollutants

Parameters	Permissible limits by various regulatory agencies				
	Food and Agriculture Organization of United Nations [48]	World Bank Group [49]	European Commission [50]	China [51,52]	ZDHC Programme [53]
Temperature (°C)	40	<3[a]			35
pH	5–9	6–9		6–9	
Colour (Pt-Co)	Not objectionable	50			50
COD (mg/L)	120	160	30–300	100–200	150
BOD_5 (mg/L)	40	30		25–50	30
TSS (mg/L)	35	50	5–60	60–100	50
Copper (mg/L)	0.5	0.5	0.05–0.5	0.5	1
Chromium (mg/L)	0.05	0.5	0.01–0.15		0.2
$N–NH^{4+}$ (mg/L)	1	10		12–20	10
Oil & grease (mg/L)	10	10			10
Zinc (mg/L)	2	2	0.1–2	2	5
Chloride (mg/L)	750				
Sulphate (mg/L)	750				
Nitrite (mg/L)	1				
Nitrate (mg/L)	10				
Sodium (mg/L)	200				
Cadmium (mg/L)	0.01	0.02	0.01–0.05		0.1
Cobalt (mg/L)	0.05	0.5			0.05
Molybdenum (mg/L)	0.01				
Sulphide (mg/L)	0.002	1		1	0.5

[a]temperature increase

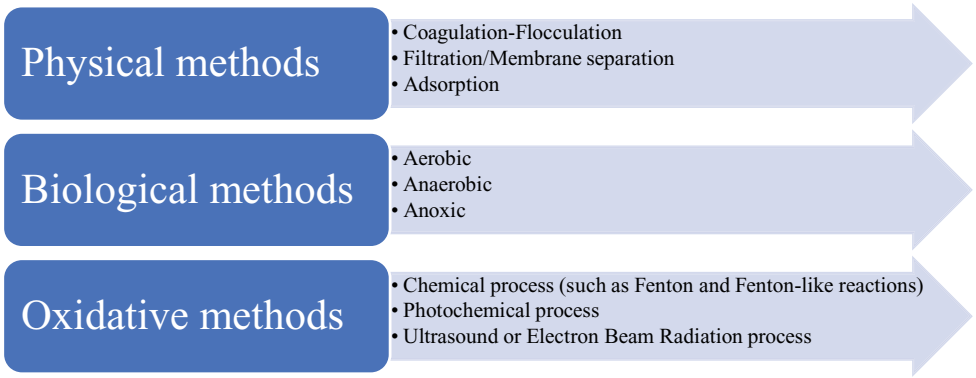

Fig. 2. Various treatment methods for treating textile wastewaters.

that it can produce biogas during the treatment of wastewater, which can be used for energy production. However, the biologically treated wastewater still requires further treatment to meet the discharge standards as the biological technology cannot remove non-biodegradable contaminants present in wastewater.

Advanced oxidative processes can be used under ambient or near-ambient conditions (though either at low or high pH values) and can effectively treat both dyes and pesticides. These methods involve using strong oxidants such as hydrogen peroxide and ozone to produce hydroxyl species [73], which react vigorously with dyes and suspended (organic or inorganic) matter to degrade them. Fenton process is another way of using advanced oxidation techniques to treat wastewater. In this method, the reaction between ferric and hydrogen peroxide is exploited to promote the oxidation of pollutants. Furthermore, these oxidants are also combined with ultraviolet (UV) radiation or ultrasonication (US) to accelerate the production of hydroxyl species and treat textile wastewater. A major disadvantage of these methods is the operating cost of the process, as most of the oxidants have a significantly short lifespan in the

wastewater environment, whereas the formed precipitates/solids (produced after the oxidation process) also require secondary treatment.

Filtration or membrane separation is an emerging technology for treating textile wastewater. Membranes offer the advantage of simultaneously removing both the organic and inorganic pollutants present in wastewater, thus reducing color, BOD and COD of the wastewater. Various membrane techniques used for textile wastewater include membrane bioreactors (MBRs), ultrafiltration (UF), nanofiltration, reverse osmosis (RO) and membrane distillation (MD). Each of these has its own pros and cons. Despite their low foot-print and hydrodynamic advantages, membranes suffer from fouling, low throughput volumes, high energy consumption and the generation of a secondary concentrated wastewater stream [74].

3. AOPs for treating textile wastewater

AOPs refer to technologies that treat recalcitrant organic contaminants present in wastewaters using highly reactive radical species, which act as oxidants while being assisted by light, catalyst, ultrasonic and/or thermal input. AOPs include ozonation, Fenton process, photochemical oxidation, EO, UV/H_2O_2, UV/O_3, photocatalytic oxidation, and sonolysis [75]. Among various reactive radical species, hydroxyl radicals have significantly high oxidation potential and are used as a secondary oxidant because they are produced from a primary, less reactive oxidative species, such as hydrogen peroxide or ozone. Due to their high redox potential, hydroxyl radicals can react with refractory compounds present in textile wastewater and convert them to CO_2, water and inorganic ions through hydroxylation or dehydrogenation. Recently, combinations of two or more AOPs have been used to enhance the treatment of various contaminants present in wastewater.

3.1. Homogenous AOPs

Ozonation is the most popular homogeneous AOP and involves O_3 and $^{\bullet}OH$ species. Ozonation can be combined with H_2O_2 or UV or both and can work both at low and high pH values. The oxidation potential of ozone is around 2.07 eV. In wastewater, under basic pH conditions, O_3 breaks down into $^{\bullet}O_2^-$ and $^{\bullet}HO_2$ radicals, which further form two $^{\bullet}OH$ that react with refractory chemicals to decompose them.

Somensi et al. [76] studied the ozonation of real textile wastewater and reported that, at a pH of 9.1, an ozone flow rate of 20 g/h reduced the COD and color by 25.5% and 67.5%, respectively, whereas the corresponding values for the pH of 3 decreased to 18.7% and 40.6%, respectively. Chu et al. [77] compared the performance of a microbubbler with that of a conventional bubbler contactor and found that microbubbler could remove 70% of COD in only 200 min, whereas the conventional bubble contactor needed 280 min for only 50% removal of COD. Similarly, they also reported that 80% of color was removed in 140 min using microbubbler device, whereas, for the same removal rate, the conventional bubbler needed 280 min. Their work highlighted the need for appropriate unit design for AOPs, which can enhance the overall efficiency of the process. Tehrani-Bagha et al. [78] used ozonation to decolorize textile wastewater containing anthraquinone dye (C.I. Reactive blue 19) and found that the initial pH of the solution does not affect the ozonation process. After 90 min, reductions of 55% and 17% in COD and total organic carbon (TOC) were observed using 800 mg/L of ozonation, respectively. Gharbani et al. [79] studied the removal of Congo red dye in textile wastewater using ozonation and found that complete decolorization (of 60 mg/L Congo red) occurred after 45 min with the ozone concentration and flow rate of 13.6 mg/L and 23 mL/sec, respectively. In addition, approximately 80% and 42% COD and TOC were removed at the pH of 11, respectively, whereas changing the pH of the system did not have any significant impact on the performance of ozonation. Overall, ozonation has the advantages of providing simultaneous treatment of color and organics, having a smaller footprint (easy installation) and producing no sludge in the system. However, the operational cost of ozonation is high due to the significant consumption of electrical energy.

Fenton or Fenton-like AOPs have received significant interest to treat wastewaters containing aromatic hydrocarbons and colored dyes. The rate of photo-Fenton reaction increases due to the formation of high-valence Fe oxidants in the presence of UV irradiation and H_2O_2. The overall efficiency of the process depends on various process parameters, such as system pH, H_2O_2 dosage, irradiation time and wavelength, and temperature. Generally, the performance of Fenton and Fenton-like AOPs is the highest under acidic pH values (2–5). However, the generation of metal-containing sludge, which requires further treatment, and the maintaining of lower pH values make these processes capital intensive. The use of sulfate-based AOPs (Persulfate (PS) and peroxymonosulfate (PMS)) has somewhat addressed this problem, as these processes work at near-neutral pH values and require shorter treatment time to produce a certain treatment efficiency. However, the capital and operating expenditures of processes employing sulfate radicals are high due to the expensive nature of these oxidants, indicating that sulfate-based AOPs are not economically feasible as compared to other counterparts [80,81].

Table 4 presents the operating parameters and performances for various homogeneous AOPs applied to various types of textile wastewaters. Homogeneous AOPs also employ various combinations of different processes, such as UV/H_2O_2, UV/Fe^{2+}, UV/H_2O_2/Fe^{3+}, H_2O_2/Fe^{2+}, and H_2O_2/Fe^{2+}/US. The most common of these processes involve using UV irradiation along with the photoproduction of Fe^{3+} ions, which are reduced to Fe^{2+} while generating new $^{\bullet}OH$ radicals from H_2O_2. The results presented in Table 4 show that the use of a particular homogeneous AOP depends upon the kind of wastewater that is needed to be treated. This arises from the complexity due to the diverse nature of pollutants present in textile wastewaters. Although ozonation has shown better performance under basic conditions, the optimization of the concentration, flow rate and equipment design for efficient use of ozonation are the major areas to be studied in this field. With regards to the use of Fenton and Fenton-like reactions to further aid the treatment process, the biggest challenge is the maintenance of pH value, as these reactions work optimally under a certain pH environment. Nevertheless, the biggest challenge

Table 4
Various strategies, operating parameters and performances of homogenous AOPs

Type of wastewater	Strategy	Operating parameters[a]	Performance	References
Real textile wastewater (pilot study)	Ozonation	pH: 3, 9.1; ozone: 20 g/h	COD removal: 25.5%; color removal: 67.5% (both at pH of 9.1); lower efficiency at pH of 3	[76]
Real textile wastewater	Ozonation	Ozone: 0.02–1.5 L/min; ozone conc.: 132 mg/L; comparison of a microbubbler and conventional bubble contactor	COD removals of 70% and 50% after 200 min (microbubbler) and 360 min (conventional bubble contactor)	[77]
CI RB 19 containing wastewater	Ozonation	Ozone conc.: 25 mg/m^3 and 55 mg/m^3; RB 19 conc.: 100–800 mg/L	COD and TOC removals of 55% and 17%, respectively	[78]
CR containing wastewater	Ozonation	Ozone: 23 mL/s; ozone conc.: 13.6 mg/L; Congo red conc.: 60 mg/L; pH: 3, 7, 11	COD and TOC removals of 80% and 42%, respectively, achieved after 45 min	[79]
Real textile wastewater	Ozonation followed by nanofiltration	Ozonation time: 30 and 60 min.; pH: 5–10; DK-5 (Osmonics Inc., USA.) and NF-100 (Dow Chemicals, USA) membranes	COD and turbidity reductions of 57% and 95% (respectively) achieved after 60 min	[82]
MB containing wastewater	Ozonation	pH: 2–12; dye conc.: 50–600 mg/L; ozone conc.: 4–24 g/m^3	COD reduction: 64.96%; decolorization in 28 min at a pH of 12	[83]
Real textile wastewater containing reactive dyes	Ozonation	T: 35°C; pH: 9.0; COD: 220 mg/L; BOD: 72.7 mg/L; reaction times: 5–360 min	Decolorization of 56.82% and 92.20% after 120 and 360 min, respectively	[84]
Red 198, Blue 21, Black 31 and Orange 107 containing wastewater	Ozonation	Dye conc.: 250–1,000 mg/L; ozone: 52.5 mg/L; reaction time: 60 min	Complete decolorization of Blue 21, Red 198, and Black 31 achieved in 30 min, while Orange 107 took 45 min	[85]
Bamboo industry wastewater	Ozonation	Ozone: 3.15 g/h; ozone conc.: 52.5 mg/L	Decolorization, COD and TOC removals of 95%, 56% and 40%, respectively, achieved after 25 min	[86]
Reactive Black 5 containing wastewater	Ozonation	T: 20°C; ozone conc.: 2 mg/L; dye conc.: 50 mg/L	Complete decolorization achieved within 15 min	[87]

Anthraquinone dye (C.I. Reactive blue; RB 181) containing wastewater	UV/H_2O_2	*T*: 20°C; H_2O_2 conc.: 100–4,000 mg/L; dye conc.: 50–250 mg/L; pH: 2–7; UV exposure time: 20 min	Complete decolourization achieved *T*: 20°C; H_2O_2 conc.: 500 mg/L; dye conc.: 100 mg/L; pH: 3; UV exposure time: 20 min	[88]
Azo dye RG19 containing wastewater	UV/H_2O_2	pH: 2–10; H_2O_2 conc.: 2.5–120 mM; intensity of UV radiation: 1,500 μW/cm^2; initial dye conc.: 0.0634 mM	Complete decolorization within 20 min. Around 63% TOC removed within 90 min	[89]
R6G containing wastewater	UV/H_2O_2/Fe^{3+}	pH: 2.3–4.7; H_2O_2 conc.: 27–173 ppm; Fe^{3+} conc.: 1.5–30.6 ppm	Around 70% mineralization achieved in 30 min	[90]
Real textile wastewater	Fe^{2+}/H_2O_2/US	pH: 2.0–4.5; Fe^{2+} conc.: 0.05–0.225 g/L; H_2O_2 conc.: 1.1–2.8 g/L; reaction time: 15–180 min	99% decolorization and 77% COD removal were achieved	[91]
Azo dye Reactive Magenta MB containing wastewater	H_2O_2/Fe^{2+}	Fe^{2+} conc.: 0–500 mg/L; initial H_2O_2 conc.: 0–12 g/L; pH: 2–3.5; initial dye conc.: 10–150 mg/L	96.7% of removal was achieved	[92]
Micropollutants in simulated as well as real wastewater	UV/PS/Fe^{2+}, UV/PMS/Fe^{2+}	PS and PMS conc.: 0.05–5 mM; pH: 7.1–7.2; Fe^{2+} conc.: 1:1 molar ratio to PS or PMS; UV-C contact time: 2.8–28 s; UV-C dosage: 5.7–57 J/L	62% organic matter mineralized for the dosage of 0.5 mM of UV-C/PMS/Fe^{2+} for UV-C contact time of 9 s and dosage of 19 J/L	[93]
Diatrizoate thyroxine containing wastewater	UV/PS	pH: 7.4; temperature: 21°C; PS conc.: 1 mM	100% removal of diatrizoate thyroxine achieved	[94]
Anatoxin-a containing wastewater	UV/PMS	Wavelength: 260–290 nm; PMS conc.: 0.15 mM; pH: 6.4; temperature: ambient	In 10 min, 98.6% of the anatoxin-a was removed	[95]
1,1,1-trichloroethane and 1,4-dioxane containing wastewater	US/PS	US power: 100 W; PS conc.: 1.5 mM; pH: 7; temperature: 15°C	After 2 h, 100% 1,1,1-trichloroethane and 60% dioxane were removed	[96]
4-chlorophenol containing wastewater	US/PMS/nanoscale zero-valent iron (nZVI)	pH: 3; nZVI conc.: 0.4 g/L; PMS conc.: 1.25 mM	In 30 min, 95% of 4-chlorophenol was removed	[97]

[a]All concentrations (conc.) in these columns refer to initial concentrations. Ranges mean that those parameters were varied within the lower and upper limits of the range.

to homogeneous AOPs comes from the metal-containing sludge that is produced during the treatment process.

3.2. Heterogenous AOPs

In heterogenous AOPs, generally, a photocatalyst is used in the presence of UV irradiation to degrade pollutants and color present in textile wastewaters. The catalysts act as semiconductor materials with a certain bandgap. When the energy of irradiated UV source exceeds the bandgap of these photocatalysts, holes and electrons are generated, both of which react with the aqueous reaction media to produce highly active •OH and peroxide radicals that increase the overall rate of heterogeneous photodegradation. Various photocatalysts including ZnO, TiO_2, ZrO_2, and ZnS have been used in heterogeneous AOPs. Among these, ZnO and TiO_2 are the most widely used photocatalysts, are easily available, cheap, non-toxic and can work under ambient-to-mild conditions. Furthermore, in some previous studies, ZnO has produced better results than TiO_2 or ZnS under similar operating conditions, which is due to wider band gaps of TiO_2 and ZnS [98]. The overall efficiency of heterogeneous AOPs depends mainly on the dosage of photocatalyst, type of photocatalyst, irradiation time and wavelength, and pH.

Xavier et al. [99] compared the homogeneous and heterogeneous oxidations of Magenta MB dye containing textile wastewater and reported that the homogeneous Fenton process (using $FeSO_4$) needed less amount of catalyst than the heterogeneous counterpart (using Fe_3O_4), whereas the dye removal efficiencies of the two processes were almost similar (90.9% and 86.8%, respectively). Furthermore, the processing time in the case of homogeneous AOP was less than that for the heterogeneous AOP. However, it must be pointed out that the heterogeneous process required a smaller H_2O_2 dosage, somewhat offsetting the process cost as compared to its homogenous counterpart. In addition, catalyst particles in heterogeneous AOP are in the solid phase and can be recovered for reuse in the process, whereas the homogeneous process requires separate treatment of metal-containing sludge, thus further increasing the overall cost of the homogeneous process.

In another study [100], the homogeneous and heterogeneous AOPs were compared and it was found that the removals of COD, BOD and TOC were higher in the heterogeneous process (using mesoporous activated carbon) than the homogenous process (using H_2O_2 and $FeSO_4{\cdot}7H_2O$). Although the study concluded that the heterogeneous process exhibited a smaller rate constant for pollutants' degradation than the homogeneous process at a certain temperature and within the same processing time, the overall COD removal efficiencies for the heterogeneous process were higher than the homogeneous process for various catalyst and H_2O_2 dosages, indicating that the heterogeneous process would be more economical than the homogeneous counterpart for certain process efficiency. This came with the added advantage of no sludge production when mesoporous activated carbon was used in the heterogeneous photo-Fenton process.

Valdés and Zaror [101] studied the treatment of benzothiazole containing wastewater using ozonation and activated carbon-assisted ozonation and found that the heterogeneous oxidation resulted in a reaction rate constant of around 0.0024 g/L sec, whereas the corresponding value for homogeneous oxidation was 0.0013 s^{-1}. The authors further reported that the removal rate of the pollutant was directly proportional to the dosage of activated carbon and resulted in around 83% of pollutant removal within the pH of range of 2–11 for the heterogeneous oxidation reaction. Although heterogeneous AOPs come with added advantages, the choice of a suitable AOP process depends on many factors, including the type and concentrations of pollutants in the influent and the degree of treatment needed for safe discharge of wastewater.

Table 5 presents a summary of various heterogeneous AOPs along with their operating parameters and performances for use in textile wastewater treatment. Compared to their homogeneous counterparts, these processes produce smaller amounts of sludge and better results in relatively shorter treatment time. Besides, when used in combination with UV, H_2O_2 or ozonation, the presence of these catalysts significantly reduce the amount and concentration of H_2O_2 or ozone needed to achieve a certain treatment. A major challenge in the development of heterogeneous AOPs, particularly those involving the use of UV, is to find a suitable catalyst, which has a wide enough bandgap and is not very expensive. The optimization of the amount of catalyst (often the photocatalyst) for a certain treatment efficiency is also an open area of research under this topic.

4. Membranes for textile wastewater treatment

Table 6 presents common membrane processes used for wastewater treatment and the characteristics of membranes used in these processes.

In textile wastewater treatment, microfiltration (MF) is generally used as a pre-treatment step upstream of a secondary, more stringent treatment process. This is due to the larger pore size of MF membranes, which lets dissolved solids and soluble pollutants pass through them. In an earlier study [121], textile wastewater was treated using the combinations of coagulation/flocculation/nanofiltration (NF) and MF/NF and it was found that the final permeate flux for MF-treated wastewater was higher (around 34 L/m^2 h) than the coagulation/flocculation-treated wastewater (ca. 14 L/m^2 h). The results were attributed to the superior performance of MF over coagulation/flocculation as a pre-treatment step, which reduced more of color, COD and salts than the coagulation/flocculation, thus improving the influent quality entering the NF membranes. In another study [122], mineral coal was sintered on a porous graphite support layer to form two MF membranes (pore size diameters of 0.5 and 0.8 μm) and the results for the treatment of real textile wastewater showed that the MF membrane with larger pores produced permeate flux of around 150 L/m^2 h, whereas the corresponding value for the other membrane was around 4.5 L/m^2 h. Furthermore, both the membranes had the same removal efficiencies for color and turbidity (87% and 89%, respectively), however, the membrane with larger pores were able to remove more COD (59%) than the other membrane (48%). It is worth mentioning that the membrane with a larger pore size achieved lower salinity removal (28%) than the other membrane (33%).

Table 5
Various strategies, operating parameters and performances of heterogeneous advanced oxidation processes

Type of wastewater	Strategy	Operating parameters	Performance	References
Real textile printing wastewater	UV/H_2O_2/TiO_2	TiO_2 conc.: 0.4–0.8 g/L; pH: 3, 9; H_2O_2 conc.: 3%, 9%; UV exposure time: 60 and 120 min.	Reductions in COD and TOC of 58%, and 48% respectively	[102]
MR73 azo dye containing wastewater	Photo-Fenton (UV/H_2O_2/Fe^0)	pH: 3; initial dye conc.: 0.05 mM; initial H_2O_2 conc.: 2.5 mM; iron dosage: 0.11 g; *T*: 25 °C	Decolorization of 99% within 15 min. Around 85% COD removal within 3 h achieved	[103]
Real textile wastewater (pilot scale)	UV/TiO_2/H_2O_2, UV/H_2O_2/Fe^{2+}	pH: 10.8; *T*: 31.2°C; DOC: 382 mg/L; COD: 1,020 mg/L; BOD_5: 110 mg/L; color: lilac.	Around 98% decolorization and 89% TOC removal achieved after 7.2 and 49.1 kJ UV/L, respectively, for 100 mg Fe^{2+}/L	[104]
MB containing wastewater	H_2O_2/ferrocene (Fc)	MB conc.: 10 mg/L; pH: 4; *T*: 30°C–60°C; Fc conc.: 0.186–0.558 g/L; H_2O_2 conc.: 7.86–31.44 mmol/L.	Use of 1.12% ferrocene (Fc) obtained 99.5% removal of MB and complete decolourisation in about 50 min	[105]
MB containing wastewater	H_2O_2/cobalt (Co)	MB conc.: 50 mg/L; diatomite-supported cobalt conc.: 0.005–0.01 g; *T*: 25°C; $NaHCO_3$ conc.: 25 mM; H_2O_2 conc.: 60–180 mM	Around 98% removal of MB and 70.4% removal of COD after 5 h	[106]
Synthetic wastewater containing SO 7GL, SB 71, solophenyl scarlet BNLE, solophenyl yellow ARL, SB FR, NB 98 azo dyes	UV/TiO_2/H_2O_2	TiO_2 conc.: 0.1–0.9 g/L; H_2O_2 conc.: 1–100 mmol/L; pH: 3; initial dye conc.: 50 mg/L.	Complete decolorization achieved in 240 min	[107]
Real textile wastewater and synthetic wastewater containing chemstar turq blue	UV/H_2O_2/modified iron oxide	pH: 2–5; catalyst conc.: 5–20 mg/L; H_2O_2 conc.: 50–200 mM; UV power: 0–24 W;	85% dye and 62% COD were removed	[108]
Textile wastewater containing a synthetic dye	UV/H_2O_2/TiO_2	H_2O_2 conc.: 0.25–5 g/L; hydraulic retention time: 0–120 min; pH: 3.	94% TOC removal achieved in 45 min	[109]
Reactive Black 5 containing wastewater	UV/H_2O_2/foundry sand (FS) and fly ash (FA)	Irradiation time: 0–120 min; FS conc.: 0.1–1 g; H_2O_2 conc.: 0.88–8.82 mM; pH: 2–7; ash: 0.05–1 g; ash/sand: 0.08–1	For FS: around 90% of degradation and decolourization achieved in 70 and 45 min, respectively; For FA: around 90% degradation in 30 min, and 93% decolourization in 15 min achieved	[110]
Rhodamine B (RhB) containing wastewater	UV/H_2O_2/$H_3PW_{12}O_{40}$@C (PW_{12})	PW_{12} dosage: 10%–50% (w/w); pH: 2–10; H_2O_2 conc.: 0–2 g/L.	Around 94.6% RhB was decolourized in 70 min	[111]
Reactive Red 120 and RR 198 containing wastewaters	UV/H_2O_2/soy meal hull activated carbon (SMHAC), UV/H_2O_2/SMHAC/TiO_2	Irradiation time: 0–75 min; pH: 3–10; dye conc.: 100–250 mg/L	Around 80% degradation of each dye was achieved within 75 min using UV/H_2O_2/SMHAC/TiO_2	[112]
Phenol containing wastewater	PS/cubic mesoporous carbon (CMK)	CMK conc.: 0.2 g/L; PS conc.: 6.5 mM; temperature: 25°C	In 20 min, 100% phenol removal was achieved	[113]
RhB containing wastewater	PMS/Cu/ZSM-5	Cu/ZSM-5 conc.: 1 g/L; pH: 7; PMS conc.: 0.60 g/L; temperature: ambient	95% of 50 mg/L Rhodamine B removed in 1 h	[114]
RR M-3BE containing wastewater	PMS/Fe@ACFs	PMS conc.: 0.05–1 mM; RR M-3BE conc.: 50 μM; g/L; pH: 3–9; temperature: 50°C	In 15 min, more than 99% of RR-3BE was removed with 2 g/L Fe@ACFs	[115]

Table 6
Common membrane processes and corresponding membrane characteristics for wastewater treatment

Process	Pore size (nm)	Transmembrane pressure (TMP) (bar)	Permeability (L/m² h bar) [116]	Targeted pollutants	References
Microfiltration	100–10,000	0.1–2	>50	Bacteria, suspended solids and colloids	[117]
Ultrafiltration	5–200	1–7	10–50	Macromolecules, proteins and viruses	[118]
Nanofiltration	1–2	5–20	1.4–12	Salts (multivalent ions), dyes and lactose (sugars)	[119]
Reverse osmosis		10–100	0.05–1.4	Monovalent ions	[120]

In recent years, MD, which is a non-pressure driven process, has received significant interest with regards to textile wastewater treatment [123,124]. In particular, the zero liquid discharge (ZLD) technology has gained plenty of interest as, in this process, the water treatment plant does not discharge any effluent to water bodies, thus completely eliminating the environmental pollution associated with these effluents [125,126]. However, the use of direct contact membrane distillation (DCMD) for textile wastewater treatment has been limited to only a few academic publications [127–132]. A recent study has outlined a comprehensive overview of the development in the application of direct contact MD process to textile wastewater treatment [133]. Major challenges hindering the commercial application of MD are the fouling of membranes [134], flux decline [135], and higher energy consumption [136] of the MD process.

Significant research efforts have been devoted to developing new membranes to improve separation performance. Bousbih et al. [137] sintered natural Tabarka clay on tubular support of Wadi Melah clay to form a UF membrane and used it to treat real textile wastewater. The results showed that, for a transmembrane pressure of 3 bar, the COD, turbidity, salinity and color removals were 77.8%, 99.7%, 23.8% and 95.6%, respectively. Babu and Murthy [138] coated PVA on poly(ether sulfone) membrane to fabricate a NF membrane and used it to treat textile wastewater containing acid, reactive and disperse dyes. The results showed that, with 1 wt.% PVA, a permeate flux of 3.06 L/m² h atm was observed, whereas the maximum dye rejection was around 98%.

In recent years, metal-organic frameworks (MOFs) are incorporated in membranes to increase water purification performance. The most common MOFs used in water treatment using membranes are UiO-66 and zeolitic imidazolate framework (ZIF-8), which use 1,4-benzenedicarboxylate and 2-methylimidazolate as linkers, respectively. In this regard, UiO-66-based membranes were prepared on alumina hollow fibers using a solvothermal method and used for water purification [139]. The results showed that membranes had a good rejection for di- and tri-valent cations with the permeance of 0.14 L/m² h bar. In another work [140], commercial alumina was used as a substrate, whereas amine-functionalized MOF (NH_2-MIL-53(Al)) was used to prepare the membrane used for water purification using vacuum MD. The results showed that the hydrophobic membrane produced a flux of 32.3 L/m² h at 60°C for 3.5 wt.% aqueous NaCl feed.

Liu et al. [141] used glutaraldehyde and monomethoxy-poly(ethylene glycol) to graft hydrophilic polymer brushes on a polyamide thin-film composite RO membrane and used it for tertiary treatment of textile effluent. The results showed that the modified membranes produced a water flux of 28.9 L/m² h and salt rejection of 98.2%. Furthermore, the membrane showed consistent salt and COD rejections even after exposure to 2,000 mg/L of chlorine at 35°C for around 1 h. Parlar et al. [142] introduced a NF treatment upstream of a RO membrane and downstream an MBR to study the effect of NF pre-treatment on the overall process. They found that the introduction of NF improved water recovery by only 0.5% than the process without NF pre-treatment. However, the process with NF pre-treatment showed significantly less TDS and color, though the final COD values for both the process schemes were the same.

Table 7 presents various membranes, their characteristics and performance for textile wastewater treatment. Although membranes have shown superior treatment performance than the conventional processes, they suffer from various process-related challenges, such as comparatively low throughput, fouling and design of larger-scale modules. The advent of nanocomposite membranes has greatly increased the robustness and performance of membranes, however, more work is needed in the field of the development of suitable nanoparticles that can be produced easily and economically.

5. Integrated advanced oxidation and membrane process for wastewater treatment

In recent years, the integration of AOPs with membrane processes has received increasing attention for effectively treating industrial wastewater. Winter et al. [153] sequentially used oxidation (ozonation and UV/H_2O_2) and membrane filtration to study the overall results of the integrated process for water treatment and found that the integrated process resulted in around 92% less fouling of membrane for 50–150 kDa sized membranes than that for the standalone filtration process. The results also showed that, for less than 8 kDa MWCO membranes, the integrated process has little effect on reducing membrane fouling (1%–18%). Wang et al. [154] treated washing textile wastewater by ozonating the influent followed by treatment in biological aeration filter (BAF) with the residence time of 3–4 h and found that the combined strategies were able to remove around 62.5% of the COD, 87.5% of the turbidity and 87.5% of the color. The BAF consisted of 3–5 mm sized ceramic balls and the authors made no comment about the recyclability or life-cycle of the BAF system. Cuevas et al. [155] combined NF with individual AOPs (solar photo-Fenton, photo-Fenton like Fe(III)-EDDS complex and ozonation) and compared

Table 7
Various strategies, operating parameters and performances of membrane processes for treating textile wastewater

Type of wastewater	Strategy	Membrane characteristics	Performance	Reference
Real textile wastewater	Nanofiltration (NF); hollow fibre (HF); feed; cross-flow	Polyamide-imide based NF HF membrane; polyethylenimine functionalized outer surface; 31 m^2/m^3	pH: 7; *T*: 40°C; COD: 3,000–8,000 mg/L; 95% COD removal achieved; permeate flux of around 1.5 L/m^2 h	[143]
DR 80, DR 23, CR, RB 2 and Na_2SO_4 containing wastewater	Ultrafiltration (UF)	Tight UF membrane (UH004, Microdyn-Nadir, Germany)	More than 98.9% retention of all direct dyes in the presence of 60 g/L Na_2SO_4. Around 98% desalination and >97% dye recovery achieved	[144]
MB, CV, AR 18 and AY 36 containing wastewater	Direct contact membrane distillation (DCMD); flatsheet	Hydrophobic PVDF (GVHP with 0.22 μm and HVHP with 0.45 μm pore sizes; Durapore membrane filters, Merck KGaA, Germany) and polytetrafluoroethylene (PTFE) membranes (TF-200 with 0.20 μm pore size; TF Pall Gelman, Pall Corp., USA)	PTFE achieved average water flux of 30.33 L/m^2 h with 100% decolorization, while PVDF-0.45 achieved 19.53 L/m^2 h with 100% decolorization	[145]
RBB KN-R, cation yellow X-2RL, Reactive Black 5, RR H-E7B and Na_2SO_4 containing wastewater	UF; cross flow filtration setup	Tight UF ceramic membrane; TiO_2/ZrO_2 skin layer with pore size of 1.16 nm on porous Al_2O_3	Permeability and dye rejection of 43.5 L/m^2 h bar and 98% were achieved	[146]
Real textile wastewater	DCMD; HF arrangement; hot feed in shell side	PVDF-Cloisite 15A nanocomposite membrane; pore size of 0.088 μm	Around 95.3% color, 93.7% TDS and 90.8% COD removal achieved. Permeate flux of around 13–22 L/m^2 h was achieved	[147]
Real textile wastewater	DCMD; commercial, hydrophobic polytetrafluoroethylene (PTFE) and polyvinylidene fluoride (PVDF) membranes	PTFE membrane with 0.22 μm pores (Shanghai Minglie Membrane Co., Ltd., China) and PVDF membrane with 0.22 μm pore size (Haining Zhongli Filtering Equipment Co., Ltd., China)	Around 96% COD and 100% color removal	[148]
RB 15, RR 15, RY 145, Reactive Black 5 and Reactive orange 16 containing wastewater	NF; flatsheet	4040-TS80-TSF-sheet membrane (TRISEP Co., USA); polyamide based with non-woven fiberglass wounded fibrous support	Permeate flux of 280.8–331.2 kL/m^2 h was achieved. More than 90% rejection for all dyes and 100% removal of COD were obtained	[149]
Real textile wastewater	NF; hollow fibre configuration	m-phenylenediamine and trimesoyl chloride modified polysulfone fibers; inner diameter: 0.0005 m; outer diameter: 0.012 m	Permeate flux of around 3 L/m^2 h and dye rejection of more than 99% for all dyes were obtained	[150]
AR 87, DB 53, AB 1, Azure A, BB 9, BG 4 containing wastewater	NF; Sterlitech crossflow cell, (product from Sterlitech Corp., USA) (CF042)	NF-270 NF membrane with isoelectric point of 3.3 (Dow FilmTec is a portfolio of membranes from DuPont de Nemours, Inc., USA)	Maximum flux declines of 48.42% for negatively charged dye molecules and 51.17% for positively charged dye molecules were observed. Flux, salt rejection and dye rejection of around 120 L/m^2 h, 50% and more than 98% were achieved	[151]
Reactive Black 5 and NaCl containing wastewater (pilot scale)	INSIDE CéRAM tubular UF membrane (150 kDa; TAMI Industries, France)	NaCl conc.: 1–4 g/L; RB conc.: 100 mg/L; TMP: 1–3 bar	Dye rejection: >75%; salt rejection: ca. 40%; permeate flux: ca. 220 L/m^2 h	[152]

the performance of overall integrated processes for treating pharmaceutical wastewater. The results showed that the integrated processes needed less treatment time and lower dosages of reagents, thus making the process more efficient and economical than the stand-alone processes. Overall, the authors recommended using solar photo-Fenton-like EDDS combined with NF to treat wastewater containing low concentrations of pharmaceuticals, which was due to low requirements for initial iron concentrations, less consumption of H_2O_2 and the absence of the need to adjust the pH of the system.

Fan et al. [156] combined coagulation, ozonation and ceramic membrane UF into a single step followed by granular activated carbon (AC) filtration and found that, unlike a process without ozonation, the integrated process with the ozone dosage of 2–5 mg/L significantly reduced the membrane fouling, dissolved organic carbon (DOC), turbidity and other wastewater contaminants (removals of 64%–100%) while producing a permeate flux of around 100 L/m^2 h. The study highlights the importance of coupling various treatment methods into single units, thus significantly reducing the capital requirements for water treatment as compared to conventional methods. Panglisch et al. [157] studied various configurations including the presence and absence of ozonation and powdered activated carbon (PAC) combined with coagulations and MF or UF to treat wastewater and found that the permeabilities of as high as 800 L/m^2 h could be achieved using commercial membranes on a pilot scale. However, the optimization of process conditions was planned for future work, and therefore, no conclusive evidence about the optimum integrated process for water treatment could be reported. Moravia et al. [158] treated real landfill leachate with AOP (Fenton) followed by MF and NF and found that the permeate achieved the then discharge standards given in the legislation of Brazil except for the concentration of nitrogen. Overall, the integrated process yielded 63% COD removal, 76% color removal, and 50% humic substances under optimized conditions of pH, H_2O_2 dosage, and $FeSO_4{\cdot}7H_2O$ dosage.

Table 8 provides some examples of the integrated processes for water treatment using AOPs and membrane technologies along with the corresponding operating parameters and performances.

It is worth mentioning that these integrated processes are still in their infancy and need more research. The use of various oxidants and catalysts in combination with membranes need to be optimized. Besides, the use of photocatalysis in combination with membranes requires the use of an acidic environment, which brings the challenge of developing membranes that are more suitable to work under harsh conditions for long time periods. A techno-economic comparison of various integrated process configurations to achieve a certain efficiency also needs to be studied to find the optimal technology combination.

6. Various integrated process strategies

Based on the studies reviewed in this work, it is inferred that AOPs have commonly been combined with biological membrane reactors (secondary treatment technologies) or filtration processes to treat wastewaters. The integrated process of AOP and membrane technologies is considered to be an effective strategy for the tertiary treatment of wastewater. Tertiary treatment of wastewater consists of a single or a series of downstream processes to supplement the secondary treatment and aims at removing specific pollutants and pathogens, such as nitrogen, phosphorus, organics, metals, turbidity, chlorine and certain pathogens. There are a number of techniques employed to achieve tertiary treatment of wastewater. They include using activated carbon (granular activated carbon and powdered activated carbon), RO, forward osmosis (FO), filtration (micro, nano and ultra) and AOPs.

Fig. 3 presents various configurations that have been employed to treat wastewater using integrated processes. In general, there are five main categories (process concepts) of integrated processes, whereas one process concept consists of stand-alone membrane technology. When only the membrane technology is used, the process requires rigorous cleaning or backwashing to avoid clogging/fouling of membranes. This would also help increase the lifecycle of membrane, thus reducing the capital cost. The rest of the integrated processes mainly consist of a combination of AOPs, coagulation-flocculation, adsorption and membrane separation technologies. In recent years, MD has been combined with other membrane separation technologies to achieve near-zero or ZLD for wastewater treatment. For example, MD or MD crystallization has been separately combined with reverse electrodialysis [168], RO [169], and nanofiltration [170] to achieve ZLD for treating wastewater.

AOPs mainly help in degrading the persistent refractory organics in wastewater and remove color. Coagulation–flocculation (CF) coarsen the fines, which can later be separated using either adsorption, gravity settling or membrane separation. Some of the common coagulants and flocculants used in the integrated processes are alum/ferric chloride [171], ferric sulfate/Magnafloc LT25 [172], poly-aluminium chloride [173], ferric chloride/anionic polyelectrolyte (HIMOLOC SS120)/resin, alum/anionic polyelectrolyte Magnafloc 919 [174], alum/polyacrylamide, $Ca(OH)_2$, polyaluminum chloride/poly (acrylic acid) polymer, chitosan, and bittern [171].

Adsorption is used in integrated processes mainly to remove turbidity, whereas membrane separation can achieve separation of a range of contaminants based upon the membrane technology employed. Various adsorbents used to treat textile wastewater include activated carbon, powdered activated carbon, single-wall carbon nanotubes, multiple-walled carbon nanotubes (MWCNTs), oxidized MWCNTs, diethylenetriamine-MWCNTs, graphene oxide (GO), reduced GO nanosheets (rGONSs), graphene, GONSs, graphene nanosheet (GNS), Co_3O_4/SiO_2 nanocomposites, TiO_2, Fe_3O_4 magnetic nanoparticles, MgO, MnO_2, Go-Fe_3O_4 hybrid composite, polyvinyl alcohol and various combinations of these adsorbents [175].

It is worth mentioning that all the AOPs mentioned in Fig. 3 are not employed in a single processing step. Instead, only one or a combination of two or more are used in an integrated process. For example, "$UV/O_3/H_2O_2/Fe^{2+}$" does not employ that photolysis, photo-ozonation, photo-peroxidation and Fenton reactions take place in a single processing unit. The representation is rather generic and shows that one or

Table 8
Various strategies, operating parameters and performances of the integrated membrane and AOP processes for treating textile wastewater

Type of wastewater	Strategy	Operating parameters	Performance	References
Synthetic industrial wastewater	Microfiltration (0.7 μm) followed by H_2O_2/UV and H_2O_2/granular activated carbon	H_2O_2/TOC: 1, 5, 10; UV exposure time: 0–16.5 min	COD, TOC and suspended solids removals of 89.8%, 90.6% and 88%, respectively. Turbidity removal of 95.5% along with complete removal of *E. coli* and Orange II dye	[159]
Real textile wastewater	Ozonation followed by biological aeration filter (BAF)	Ozone: 30–45 mg/L; retention time: 3–4 h	COD removal: 62.5%; turbidity: 87.5%; color: 87.5%	[154]
Dyestuff wastewater (pilot scale)	Catalytic ozonation coupled with Ti–Mn coated tubular ceramic membranes (100 nm)	Ozone dose: 1.5–3 mg/L; operating pressure: 0.1–0.25 MPa	COD removal: >90%; suspended solid removal: >99%; complete removal of *E. coli*; permeate flux: >95 L/m^2 h	[160]
Norfloxacin, ofloxacin, roxithromycin and azithromycin (antibiotics) containing wastewater	NF followed by UV/O_3 treatment	NF time: 0–500 min; AOP time: 0–30 min; feed flow rate: 0.35 m/s; temperature: 20°C; pressure: 0.2 MPa	Antibiotics, DOC and acute toxicity removals of >87%, 40%, and 58% achieved, respectively	[161]
Pharmaceutical (etodolac containing) wastewater	Fenton oxidation (H_2O_2/Fe^{2+}) followed by NF (FM NP010; flatsheet; Microdyn-Nadir GmbH, Germany; 1,000 Da)	Fe^{2+} dosage: 0–0.1 M; pH: 3; H_2O_2 dosage: 0–5 M	COD removal of 82%; etodolic removal of >99.5% achieved. Permeate flux of around 45 L/m^2 h achieved	[162]
CR containing dye wastewater	Photocatalytic oxidation (Ni support coated with TiO_2) followed by UF (Haitao; hollow Fibre; hydrophilic polyacrylonitrile) and RO (polyimide)	pH: 2–11; irradiation time: 0–150 min; RO pressure: 0.4–0.6 MPa	Optimum pH and irradiation time for photocatalysis were 4 and 90 min, respectively. Optimum pH for UF and RO were 10 and 4, respectively. Optimum RO pressure: 0.4 MPa	[163]
C.I. DR 73 containing textile wastewater	Photocatalysis (TiO_2 aeroxide P25) combined with MF (hollow fibre; Polymem Co., France)	Irradiation time: 0–180 min; pH: 3–10; photocatalyst loading: 0–2 g/L; Dye conc.: 50–100 ppm.	Around 90% and 98% removal rates for dye and COD achieved, respectively	[164]
RR 180 and Reactive Orange 16 containing textile wastewater	Photocatalysis (ZnO and TiO_2) followed by DCMD (polypropylene (PP), polytetrafluoroethylene (PTFE) and polyvinylidene fluoride (PVDF); 0.22 μm pore size)	Photocatalyst loading: 0.25–1 g/L; distillation temperature difference: 25°C–35°C; feed flow rate: 210–665 mL/min	Distillate flow rate sequence: PP > PTFE > PVDF; COD removal efficiency: PTFE > PVDF > PP. Maximum COD removal: ca. 55%	[165]
Real textile wastewater	Photocatalysis (ZnO and TiO_2) combined with flatsheet UF (polyethersulfone UF membrane; 0.04 μm pore size; membrane photocatalytic reactor) and RO (polyimide; X20; DuPont de Nemours Co., Ltd., USA)	Photocatalyst loading: 0.25–1 g/L; *T*: 25°C	Permeate flux of around 40 L/m^2 h; 100% color removal; 30%–55% COD removal; hydraulic retention time: 6 h. With RO, COD removal of 88% achieved	[166]
AR 1 reactive dye containing wastewater	Photocatalysis (UV/TiO_2; AV-01; PRECHEZA, Czech Republic) combined with HF microfiltration (polypropylene HF; ZENA, Czech Republic)	Catalyst loading: 0.01–2 g/L; dye conc.: 15–75 mg/L; pH: 3–11; filtration time: 0–240 min	Permeate flux: 40 L/m^2 h; complete decolorization achieved in 350 min; around 75% COD removed in 600 min	[167]

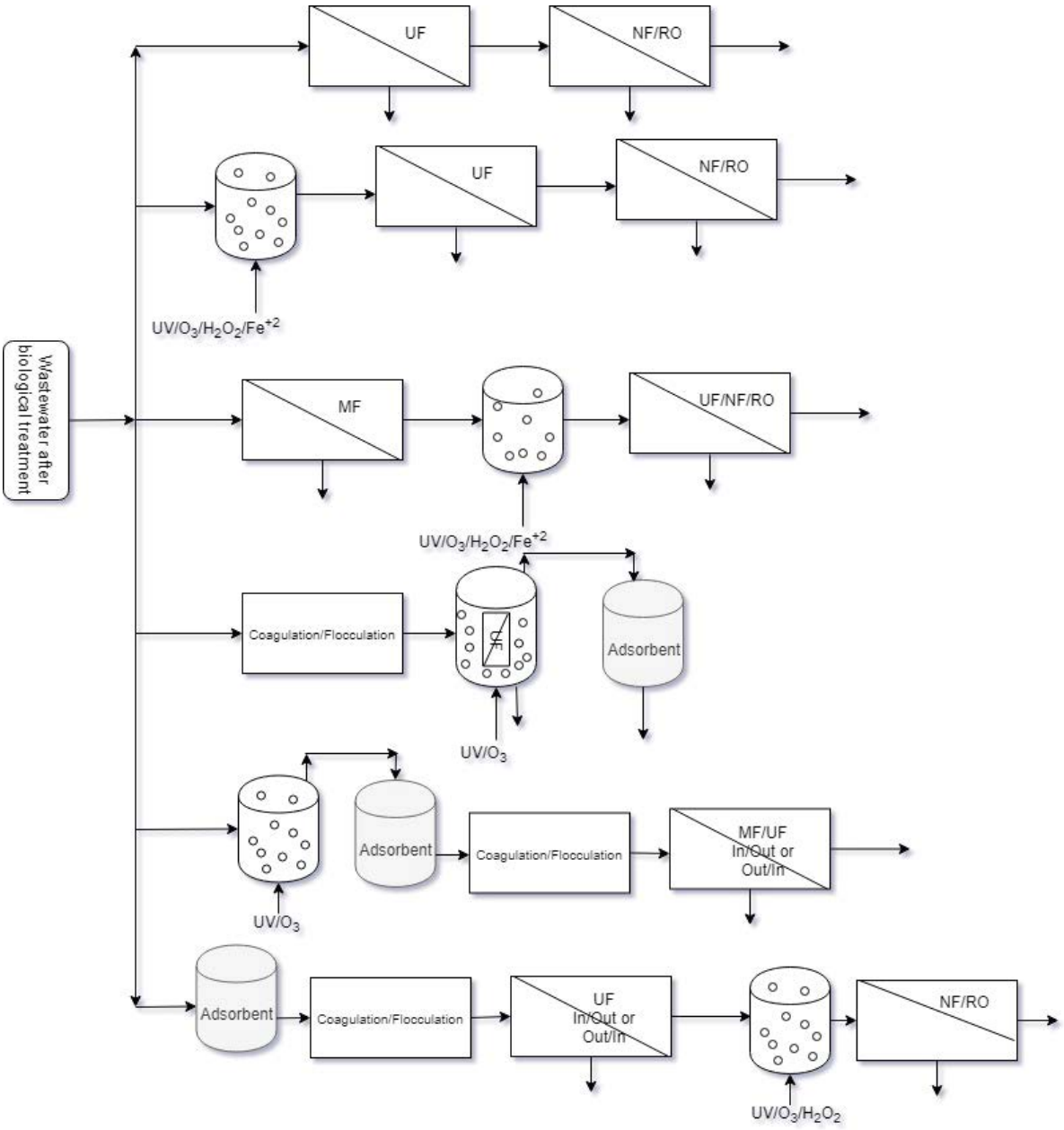

Fig. 3. Various integrated process strategies for treating wastewater.

a combination of two or more of these processes has been used with other separation techniques to achieve water purification. The same is true with regards to the terminology of "UF/NF/RO", as shown in Fig. 3. Similarly, it should be noted that different types of adsorbents are used in the integrated process for textile wastewater treatment. The effort has been made to keep Fig. 3 as generic and simplified as possible.

Although various integrated AOP – membrane-based process concepts have been proposed for tertiary treatment of wastewaters, studies on their detailed techno-economic comparison are still scarce in the literature. In a previous study [176], authors performed a techno-economic analysis on various advanced treatment technologies for treating olive mill wastewater (OMW) and found that the photo-Fenton (Fe^{2+}) process produced the product stream with the least phytotoxicity as compared to those of UF/Jet-loop reactor (Jacto MBR) and integrated UF/NF processes. With regards to the capital and operating expenses, the authors of the same study found that, for a period of 10 y and treating 1 m^3 OMW/d, the total expense of running Jacto MBR, photo-Fenton (Fe^{2+}) and integrated UF/NF processes were 194, 424, and 307€, respectively. However, the authors highlighted that the integrated UF/NF process could separate valuable hydroxytyrosol and tyrosol through concentrate stream, which could bring in a profit of 370–610 €/m^3 of

OMW treatment, thus making the membrane process more beneficial to the other two. In another study [177], the economic comparison between UF pre-treatment and conventional pre-treatment (in-line coagulation and 2-stage sand filters) for a large scale seawater reverse osmosis (SWRO) plant was conducted, and it was reported that the ratios of specific investment costs (former to latter) and operational costs were 1.01 and 0.98, respectively, indicating that integrated membranes processes are financially competitive to conventional treatment technologies. It is worth mentioning that the authors of the same study [177] did not consider land acquisition cost in the analysis. It is well known that membranes incur a smaller land footprint (around 60% smaller) as compared to conventional technologies, which would have further reduced the investment cost for an integrated membrane process [178,179].

Integrated AOP – membrane processes also harbor the advantage of using less chemicals and thus producing smaller volumes of waste/by-products. It is also well-known that conventional water treatment processes involved the consumption of large amounts of chemicals during coagulation/flocculation/sedimentation, and this results in the production of large amounts of sludge (and by-products) that requires further treatment [180,181]. If large amounts of chemicals are used, the treatment of resulting sludge might represent a large proportion of the overall expense of water treatment plants. The integrated membrane processes require lesser chemicals, though they may need chemical cleaning to avoid severe fouling. In addition, the final concentrate streams might contain some harmful chemicals, used in the pre-treatment and cleaning processes [182,183]. To the best of our knowledge, no study has yet been conducted that compares the chemical usage and waste production from the integrated membrane and conventional water treatment processes. However, this is comprehendible that membranes require lesser use of chemicals (like the removal of species in the pre-treatment section does not involve using chemicals, rather involves using membranes) than the conventional processes, and this will further reduce the use of cleaning chemicals for downstream membranes, thus reducing the overall cost of chemicals and the production of waste/by-products in the integrated membrane process for water treatment [184].

7. Challenges for a robust integrated process

Integrating AOPs with membrane separation is the right direction to harness the benefits of both technologies in a combined way. However, there are still a number of challenges, which remain to be resolved for even pilot testing of the integrated processes.

Generally, membranes (with the exceptions of MD and forward osmosis) are a relatively mature technology than AOPs, and therefore, have less challenges for applications in an integrated process. Nevertheless, the main issue with membranes is the fouling of membranes and the removal of cake material for continuous operation of the process. With a variety of contaminants present in textile wastewater (Tables 2a and 2b), the clogging of membranes would pose a challenge for a large-scale integrated process. A number of studies have addressed these issues, proposing various solutions, however how these solutions fair on a larger scale in an integrated process for textile wastewater application is still to be studied. In addition, for emerging membrane technologies, such as MD and forward osmosis, the fabrication and development of large-scale membrane modules for pilot testing is another critical aspect that needs further attention. With the development of more advanced membranes on a laboratory scale, technology needs to keep pace to upscale the production of these membranes in a cost-effective manner, so that they could be used on a larger scale.

When considering the homogeneous AOPs, the irradiation (UV) struggles to penetrate turbid waters, thus making it significantly inefficient to degrade contaminants. In order to combat this issue, photolysis is aided with other treatment methods. Simple ozonation (O_3) can be used to treat refractory wastewater, however, the kinetics of degradation using ozonation are orders of magnitude slower than with hydroxyl ions. Therefore, the process is sped up using a combination of ozonation and hydrogen peroxide to treat wastewaters. However, with such techniques, the cost of chemicals becomes a key factor, determining the overall cost of the process. Although some researchers have studied the *in situ* productions of O_3 and H_2O_2 to minimize the overall cost of the process, the optimization of the process has not yet been reported, and therefore, remains a key challenge for pilot scale testing of an integrated process.

Another key challenge is the reduction in the production of harmful by-products during wastewater treatment using AOPs. Excess ozonation can produce toxic products, which might remain within the water body, thus posing a danger to human and aquatic life. Similarly, the presence of unused H_2O_2 in treated wastewaters might result in the growth of pathogens, which again contribute to the toxicity of water bodies. Due to these reasons, optimization of the quantities of ozone and hydrogen peroxide for wastewater treatment is a critical factor for the efficient treatment of wastewater.

The homogeneous Fenton process suffers from the problem of iron-containing sludge, which requires further treatment. Although there have been some studies on using this sludge as a Fe source, the mere need for further treatment of sludge makes this process cost-intensive. Once the sludge treatment or recycling process is integrated into the overall Fenton process for wastewater treatment, the technology would become more favorable for use on a larger scale. Another drawback of Fenton's process is that the reaction takes place at low pH values (Table 4). The maintenance of low pH is not a critical issue, however, for a scaled-up process, low pH values will play a crucial role in the material design of reactors and pipelines.

With regard to the heterogenous AOPs, the biggest challenge is to find a suitable catalyst (second phase) to treat wastewater. Most of the catalysts, which have been used in wastewater studies, are expensive and not suitable for large scale applications. To further aggravate the situation, these catalysts undergo leaching, thus making the overall process more unfavorable for a large-scale application. Besides, if the catalyst is used in the form of a slurry, an extra treatment step is needed to recover or recycle it, which adds further capital and operating expense to the process. In addition to the introduction of a secondary phase (solid), which already complicates the handling of unit operations, catalyst

deactivation or poisoning is also a key issue needing further research. Such deactivation or poisoning might happen due to a reaction with intermediate products, by-products or adsorption on one or more of these products. In order to make a heterogenous process more favorable on a larger scale, further study is needed to find a cost-effective and suitable catalyst for treating textile wastewaters.

A suitable design of a reactor for a stand-alone or integrated AOP process is another key challenge for a large-scale application of the technology. Currently, there are no studies detailing the design of an upscaled reactor involving AOPs. The optimization of various parameters, such as uniform penetration of UV, amounts of ozone/H_2O_2 for a reasonable mass transfer operation, and retention times still need to be studied. Varying discharge concentrations of pollutants in wastewaters, originating from the same textile factory, also undermine efforts for designing a more robust reactor for this purpose.

Currently, some ground-breaking work has been conducted to study the combination of membrane processes only, however, the choice of a suitable integrated AOP – membrane separation process for a certain wastewater treatment application and its techno-economic analysis is still scarce in the literature. The literature has a significant gap for the techno-economic or life-cycle assessment of water treatment processes employing various integrated AOP-membrane separation processes.

Lastly but most importantly, the analysis of intermediates or products produced after wastewater treatment should be conducted to ensure that these products are not harmful to the environment or human health. Currently, the focus is only on removing or degrading the contaminants present in wastewaters, and little attention is paid to the fate of products produced during the treatment process. Such studies would prove vital for the upscaled operation of an integrated process involving both AOPs and membrane separation.

8. Conclusions

In this work, a review of some recent studies on AOPs, membrane separation and combined AOP-membrane processes for treating textile wastewater is conducted. Homogeneous and heterogeneous AOPs mainly consist of ozonation, H_2O_2, Fenton, Fenton-like and their combinations with UV irradiation and the US. Ozonation and UV/H_2O_2 are the most common processes used to treat textile wastewater, even on large scale. However, the uniform penetration of UV and optimization of the amounts of oxidants are the key factors needing further research for more robust large-scale applications. Fenton reaction is widely studied in the literature, whereas more work is emerging on the benefits and use of heterogeneous Fenton or Fenton-like processes, such as employing sulfate radicals. Heterogeneous AOPs using transition metal-based catalysts have been reported to achieve higher kinetics in shorter treatment times, and generate a lesser amount of sludge than the homogeneous Fenton process. The use of (photo-) catalysts and adsorbents to treat wastewater is also being studied. However, the search and development of a cost-effective and more robust catalyst/adsorbent is an imminent research direction. An emerging technology for treating textile wastewater is the use of the membrane process. The development and upscaling of novel membranes is a critical issue needing further attention. Membranes containing various nanocomposites are manufactured and used on a lab-scale, however, the cost and fabrication of these membranes on a larger scale in the form of modules are still lacking research. A handful of studies have reported the integration of AOPs and membrane separation for textile wastewater treatment on a laboratory scale. These integrated processes have shown competitiveness and certain advantages over their conventional counterparts, particularly with regards to the use of chemicals and the production of waste/by-products during water treatment. However, there is still a research gap in the process integration, design of required equipment (such as reactors), continuous operation of the overall process, comparison of various treatment strategies (process concepts) in an integrated process and the upscaling of the final process. Therefore, in addition to optimizing individual processes, the main future works will include finding a feasible AOP-membrane separation process for tertiary textile wastewater treatment and performing a techno-economic or life-cycle assessment comparison with other technologies to further the use of such processes in water treatment. Once such an integrated process is realized, the next step would be to unify and integrate this process into the conventional treatment process comprising primary, secondary and tertiary treatment, and optimize it for water recovery with higher flux without compromising much on overall expenditure and water quality.

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