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Review

Remediation of textile effluents by membrane based treatment techniques: A state of the art review

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Abstract

The textile industries hold an important position in the global industrial arena because of their undeniable contributions to basic human needs satisfaction and to the world economy. These industries are however major consumers of water, dyes and other toxic chemicals. The effluents generated from each processing step comprise substantial quantities of unutilized resources. The effluents if discharged without prior treatment become potential sources of pollution due to their several deleterious effects on the environment. The treatment of heterogeneous textile effluents therefore demands the application of environmentally benign technology with appreciable quality water reclamation potential. These features can be observed in various innovative membrane based techniques. The present review paper thus elucidates the contributions of membrane technology towards textile effluent treatment and unexhausted raw materials recovery. The reuse possibilities of water recovered through membrane based techniques, such as ultrafiltration and nanofiltration in primary dye houses or auxiliary rinse vats have also been explored. Advantages and bottlenecks, such as membrane fouling associated with each of these techniques have also been highlighted. Additionally, several pragmatic models simulating transport mechanism across membranes have been documented. Finally, various accounts dealing with techno-economic evaluation of these membrane based textile wastewater treatment processes have been provided.

Keywords: Textile; Effluents; Membrane; Reclamation; Model; Techno-economic evaluation

1 Introduction

Today's world stands as a witness to the revolutionizing socio-economic impacts of various industries. Unfortunately, the development of industrial sector has whipped up certain unintended repercussions, resulting in an unavoidable trade-off between industrial progress and environmental degradation. Textile industries, for instance, are one of the largest consumers of water, dyes and various processing chemicals that are used during the various stages of textile processing. Subsequently, substantial quantities of effluents are generated, mostly consisting of spent or unutilized resources, which are not suitable for further usage. These effluents are likely to cause environmental problems if discharged without prior treatment. The wastewater obtained from the textile industry is usually rich in color, chemical oxygen demand (COD), complex chemicals, inorganic salts, total dissolved solids (TDS), pH, temperature, turbidity and salinity (Verma et al., 2012; CPCB, 2007). According to the classification suggested by Environmental Protection Agency (USEPA), textile wastes can be divided into four principal categories, namely the dispersible, hard-to-treat, high-volume, and hazardous and toxic wastes (Foo and

Hameed, 2010). Among the various complex constituents present in textile wastewaters, the dyes can be inarguably considered as the most peremptory source of contamination. The direct discharge of the coloured textile effluent into the fresh water bodies adversely affects the aesthetic merit, water transparency and dissolved oxygen content (Duarte et al., 2013; Wang et al., 2009). Besides, these dyes exhibit highly complex structure, high molecular weight and low biodegradability (Verma et al., 2012; ElDefrawy and Shaalan, 2007). This accounts for its toxic effects on flora and fauna present in the water bodies. Further, these dyes are mutagenic and carcinogenic (Wang et al., 2009). The presence of these relatively recalcitrant dyes along with inorganic salts, acids, bases and other residual chemicals in the effluent directly discharged into the sewage networks impedes the biological treatment processes (Arslan-Alaton et al., 2008). Also, the chance evaporation of these chemicals in to the air we breathe or adsorption onto human skin is capable of inducing allergic reactions (Khandegar and Saroha, 2013).

Perhaps, the greatest danger to environmental sustainability is posed by the outrageously high amount of primary water consumption by the textile sector, which has, in all probability, resulted in the depletion of available fresh water resources. The deficit in the availability of water can be gauged by the fact the currently the Indian textile industry consumes 0.2 m^3 of water per kg of textiles fabricated (Parvathi et al., 2009), while generating 200–350 m^3 of wastewater per ton of finished product (Ranganathan et al., 2007). According to the recent survey conducted by FICCI Water Mission (2011), the water demand for the industrial sector is likely to witness a rise due to the impending industrial growth as also a significant rise in population; this will probably account for 8.5 and 10.1 per cent of the total freshwater withdrawal in 2025 and 2050 respectively. Thus, a 4 per cent hike from the current 6 per cent level of the total freshwater abstraction by the industries (as per 2010 statistics) is estimated. The dwindling supply of water is hence a concomitant outcome of development of industrial sector, and is bound to bring about a declination in the performance of the textile sector owing to the aggravated paucity of water resources, or deterioration in the quality of water available. These deleterious consequences have compelled the researchers to examine the suitability of the various conventional treatment technologies for treating textile industry wastewater. The sole objective of such investigations is to devise and develop a wastewater treatment technique which is environmentally compatible, cost-effective and at the same time successful in reducing the concentration of various contaminants in the textile effluent to permissible levels, which comply with the current environmental imperatives. The effluent treatment process should also be equally adept in reclaiming the water using in textile processing to a great extent; such an arrangement is indispensable for sustainable development of the industrial sector and of the country as a whole.

Various treatment techniques are in use to mitigate the contaminant levels of textile wastewaters. Table 1 provides a broad overview of the various conventional as well as recently engineered treatment processes employed to bring about the treatment of textile effluents. However, these methods suffer from certain serious handicaps. For instance, the otherwise eco-friendly biological processes, such as the conventional activated sludge systems (Lotito et al., 2012b, 2011) or anaerobic textile waste bioremediation processes (Tűrgay et al., 2011) often lack flexibility; their respective efficiencies are adversely affected by the biologically persistent constitution of the pollutants present in the textile wastewater as well as by the diurnal fluctuation in the problem environment in terms of variation in wastewater pH, temperature or concentration of contaminants in the textile wastewaters (Kapdan et al., 2000; Oller et al., 2011). Additionally, these biological treatment methods do not bring about complete mineralization of the target dye contaminants. Hence, the toxicity of the discharged effluent is often exacerbated by the chance regeneration of the primary organic constituents of the textile dyes. This drawback severely impedes the scale-up of the biological treatment technique due to the resulting reactor instability (ElDefrawy and Shaalan, 2007; Robinson et al., 2001). The complex rheology of the textile discharge therefore entails either singular or combined application of the physicochemical methods, such as chlorination, coagulation–flocculation (Al-Ani and Li, 2012; Gao et al., 2007; Yang et al., 2013), adsorption (Mezohegyi et al., 2012) and advanced oxidation processes, such as, ozonation (Somensi et al., 2010), Fenton treatments (Karthikeyan et al., 2011), electro-Fenton methods (Yu et al., 2013), photo-Fenton oxidation processes (Punzi et al., 2012), and photoelectrocatalytic reaction (Sapkal et al., 2012), for complete degradation of the toxic textile wastewater components (Álvarez et al., 2013; Lotito et al., 2012a; Oller et al., 2011; Torrades and García-Montaño, 2014). The potential of the adsorption technology remains largely untapped due to the limitations posed by environment-friendly disposal of spent adsorbents, difficulty in regeneration of spent adsorbents, reduction in reactivated adsorbent efficiencies, high costs of the adsorbents and the maintenance expenses involved (Robinson et al., 2001; Verma et al., 2012). Advanced oxidation processes, such as, ozonation are not economically attractive (Ong et al., 2014). Additionally, ozonation suffers from an inconveniently short half-life and hence the technique usually exhibits inadequate decolourization efficiencies for insoluble azoics and disperse dyes, which are prone to slow reaction. Its stability, moreover, fluctuates severely with variations in temperature, pH, and salt concentrations (Robinson et al., 2001; Verma et al., 2012). The chemicals used in operations such as coagulation and chlorination not only increase the treatment costs, but also tend to amass by-products and residues in bulk; these unconsumed waste products, thereafter, evolve into sources of secondary pollutants, hence resulting in considerable declination in the recovered water quality. Furthermore, the time consuming trials involved in selecting the coagulant/coagulants suitable for a specific kind of effluent also adds to the limitations of the coagulation–flocculation technique (Chakraborty, 2010; Robinson et al., 2001). Additionally, the degradation products present in the recycled liquor, more often than not, adsorb onto the fibres during the fabrication of textiles, and tamper with the dyeing process, to the detriment of fabric quality (Schäfer et al., 2005). Also, the effectiveness of operations such as flocculation is markedly curbed by the high electrolytic strength usually observed for the textile effluents. Besides, these chemicals and by-products generate a huge volume of sludge, which makes sludge handling difficult and contributes significantly to disposal costs (Somensi et al., 2010; Yang et al., 2013). These drawbacks can be satisfactorily overcome using membrane based wastewater treatment processes, which include microfiltration, ultrafiltration, nanofiltration, reverse osmosis or hybridization of two or more of these membrane based techniques. The membrane technology is normally hailed as clean and environmentally benign technology. The inherent simplicity of the membrane technology, the provision of modular design for handling large industrial-scale feed volumes, operation under moderate temperature conditions with no phase change, and the negligible use of additives are some of the advantageous aspects of membrane based treatment techniques (Dutta, 2007). Besides, no waste by-products or secondary pollutants are usually encountered; additionally, the appreciable retention efficiencies and stability characterizing most of the membrane based processes under varying experimental environment enable easy scale up of these techniques (Dasgupta et al., 2014; Koltuniewicz and Drioli, 2008; Ong et al., 2014) These advantages hence account for the growing interest in membrane technology. However, a major downside of the membrane based processes is membrane fouling (Van der Bruggen et al., 2008). Abatement of membrane fouling problems and reduction in membrane replacement costs can be brought about by regular cleaning of membranes and appropriate selection of the membrane filtration techniques, in accordance with the textile waste stream characteristics (Cheng et al., 2012; Dutta, 2007).

Furthermore, the initial capital or start-up costs are offset by the expenses saved in terms of competent reuse of salts, sizable recovery of dyes and water (Kurt et al., 2012; Qin et al., 2007). The reclaimed water is usually characterized by low hardness making it suitable for reuse in textile facilities (Ranganathan et al., 2007). Additionally, the relatively short payback period witnessed in many membrane based techno-economical investigations has made the membrane based treatment processes comparatively more cost-effective than other energy-intensive processes, such as evaporation (Praneeth et al., 2014); the operational cost advantages over conventional treatment methods is also an added benefit. The membrane based techniques are hence currently viewed as technologically and economically lucrative options for industrial effluents treatment; the textile industry is one of the principal beneficiaries of the membrane based treatment processes (Fersi et al., 2005; Koltuniewicz, and Drioli, 2008; Marcucci et al., 2002).

Table 1 Overview of various conventional as well as recently engineered physicochemical, biological and membrane based treatment processes employed to bring about the treatment of textile effluents.

Process adopted	Effluents characteristics	Reference
Catalytic degradation (biosynthesized silver Nanocatalysts)	Methyl orange, methylene blue and eosin Y	Vidhu and Philip, 2014
Adsorption (PES/PEI nanofibrous Membrane)	Anionic dyes, Sunset Yellow FCF, Fast Green FCF, Amaranth	Min et al., 2012
Catalytic ozonation (activated carbon, ceria catalysts)	One acid azo dye, CI Acid Blue 113, two reactive dyes, CI Reactive Yellow 3 CI Reactive Blue 5.	Faria et al., 2009
Adsorption (activated carbon)	Raw textile effluent obtained from a cotton textile mill	Ahmad and Hameed, 2009
Biodegradation (facultative Staphylococcus arlettae Bacterium)	Textile azo dyes: CI Reactive Yellow 107, CI Reactive Black 5, CI Reactive Red 198, and CI Direct Blue 71	Elisangela et al., 2009
Electrolysis (anode materials: $Ti/Ru_{0.3}Ti_{0.7}O_2$; $Ti/Ir_{0.3}Ti_{0.7}O_2$; $Ti/Ru_XSn_{1-X}O_2$, with X = 0.1, 0.2 or 0.3)	Real textile effluent	Malpass et al., 2008
Electrolysis ($Ti/Ru_{0.3}Ti_{0.7}O_2$ DSA® type electrode)	Real textile effluent	Malpass et al., 2007
Electro-Fenton process	Synthetic textile wastewater (Reactive Blue 49 dye (RB49) and Polyvinyl Alcohol (PVA))	Yu et al., 2013
Photo-Fenton oxidation (Solar and UV-C irradiation, Zero-valent iron (ZVI) catalyst)	Synthetic wastewater (azo dye, C.I. Reactive Black 1 (RB1))	Grčić et al., 2012
Photodegradation (TiO_2/H_2O_2 and Sunlight)	Real textile effluents	Garcia et al., 2009
Photocatalyzed degradation (UV-C/TiO_2, UV-C/H_2O_2 and UV-C/TiO_2/H_2O_2)	Azo-dye Reactive Orange 16	Egerton and Purnama, 2014
Ozonation (Semi-batch reactor)	Remazol Red RB Remazol Turquoise Remazol Black RL Remazol golden Yellow RNL	Tabrizi et al., 2011
Ozonation (Batch reactor)	Persistent anthraquinone dye C.I. Reactive Blue 19.	Tehrani-Bagha et al., 2010
Coagulation/flocculation (CF)/ Microfiltration (MF), CF/ULtrafiltration (UF) and CF/Powdered Activated Carbon (PAC)	Simple textile effluent (dyeing processes) complex global effluents(dyeing, bleaching and washing outlets)	Harrelkas et al., 2009

The present review paper explores the degree of success achieved by several membrane based filtration processes in bringing about appreciable reduction of the various contaminants present in the textile effluents below permissible levels. This critical assessment also seeks solutions to the problems faced by membrane technology from the analyses outlined in various investigations reported in literature. Archival reports on the methods or feasible modifications applied to enhance the economic viability of these techniques have also been assayed.

2 Various stages of textile manufacturing industry: composition and characteristics of the generated wastewaters

The processing techniques used in the various textile mills can be broadly classified as wet processing and dry processing, in accordance with the properties of the effluents generated therein (Verma et al., 2012). Effluents generated in textile mills, especially in the wet processing ones vary greatly in composition and degree of toxicity, depending on the recipes of raw materials administered, specific processes in operation, the current processing stage under consideration, the machineries and equipment employed, the quality of the water used for processing and the prevalent management theory implemented to monitor water use; whereas, dry processing units mostly generate solid wastes comprising fabric rejects (US

EPA, 1996; Volmajer Valh et al., 2011). The textile industries have been unanimously proclaimed as water intensive industries. Water is mostly used for scrubbing the raw material and for steps that require flushing; desizing, scouring or kiering, bleaching, mercerizing, dyeing, washing, neutralization, and salt bath are mostly the process steps that use water. Fig. 1 demonstrates the entire chain of steps involved in textile manufacturing; stages requiring water have been especially highlighted. However, as mentioned earlier, the water consumption varies with each process scaffold. For instance, certain operations such as dyeing and print after washing are more water intensive than the others (US EPA, 1996). Furthermore, raw materials, such as wool and felted fabrics consume more water than synthetic fibres, to fulfil their high scouring needs. Additionally, different types of processing equipment demand varying water requirements in accordance with the degree of technological constraints encountered; for example, hank machines and dyeing winches are the biggest water consumers, with water consumption ranging from 0.02 to 0.03 m^3/kg. But, water consumption by batch dyeing machines for knitwear has declined from 0.03 m^3/kg to only 0.006 m^3/kg of treated material over the last four decades due to technological advancements (Volmajer Valh et al., 2011). Hence, the heterogeneity observed in the composition as well as in the ecotoxicological effects of the textile effluents discharged from varies sources can be attributed to a number of factors. These parameters include the type and dosage of the selected raw materials, which are in turn governed by the specific techniques and machineries adopted in each mill, the processing stage under consideration and desired quality to be imparted to the manufactured fabrics.

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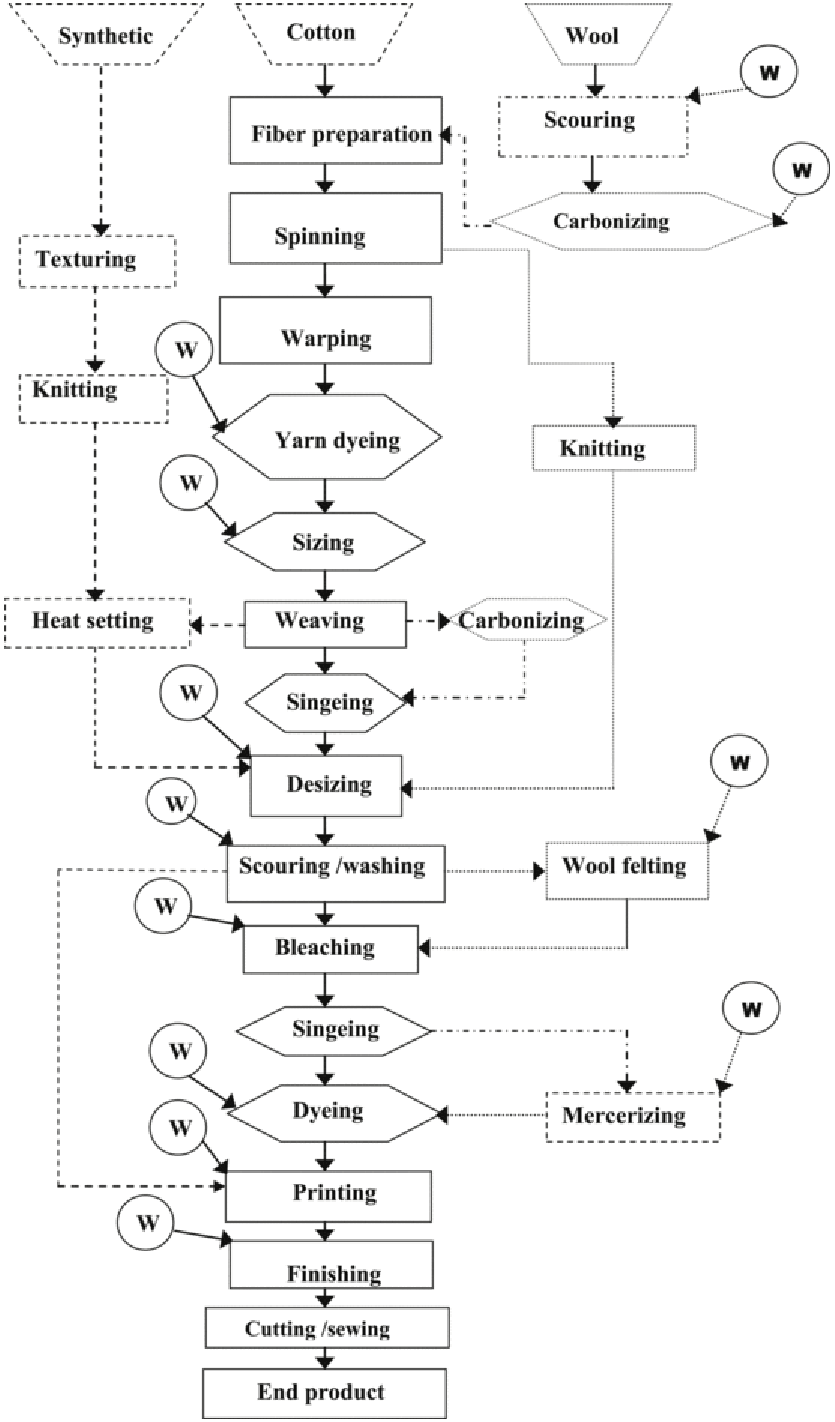

Fig. 1 Flowchart for the general steps in textile fabrication (Volmajer Valh et al., 2011).

2.1 Wastewater in textile industries

Wastewater from textile sector comprises process water, cleaning water, non-contact cooling water, and storm water (Verma et al., 2012). Major contribution to the total amount of textile wastewater generated is made by the processing stages, which include scouring, dyeing, printing, finishing, and washing (Li et al., 2012). The volume and composition of wastewater exhibit wide heterogeneity, owing to a number of factors, including the characteristic quality imparted to processed fabric, properties and chemical and physical characteristics of the applied dye, nature of special finishing if any specificity of the process, the equipment used, and the principles on which water use has been modelled. For instance, significant generators of large-volume wastes include wash water from preparation and continuous dyeing, alkaline wastewater resulting from fibre preparation and batch dye wastewater containing large amounts of residual dye, salts, acids, or alkalis, and other noxious additives in smaller amounts (Volmajer Valh et al., 2011); also operations such as dyeing, printing and finishing are mostly accountable for the deleterious rise in biochemical oxygen demand (BOD), chemical oxygen demand (COD), total dissolved solids (TDS) and total suspended solids (TSS); unusual exhibition of acidity or alkalinity is thereby witnessed, owing to the presence of toxics such as metals, salts, unused dyes, surfactants and any other organic and inorganic processing assistance such as spent solvents. Besides, residual chemicals such as batch dumps, knitting lubricants, insecticide residues, pectin, wax, disinfectants, detergents, starch-sizing agents and other organic and inorganic stabilizers obtained from operations such as bleaching and desizing, are other primary sources of biological oxygen demand (BOD) of textile wastewaters. Incidentally, desizing accounts for about 50% of the entire textile wastewater generated (Babu et al., 2007; Volmajer Valh et al., 2011). Table 2 lists the characteristic parameters of textile wastewater obtained from the various stages of textile processing.

Table 2 Characteristics of textile wastewater obtained from different stages of textile processing (data compiled from Khandegar and Saroha, 2013; Volmajer Valh et al., 2011; Ciabatti et al., 2010; Chakraborty, 2010).

Characteristics	Scouring	Bleaching	Mercerising	Dyeing	Composite	Discharge limit into public sewage Bureau of Indian Standards (BIS-3306-1955)	Maximum permissible limit for water reuse (Chakraborty, 2010)
pH	9–14	8.5–11	8–10	1.5–10	1.9–13	5.5–9.0	6.5–9.2
TDS (mg/L)	12,000–30,000	2500–11,000	2000–2600	1500–4000	2900–10,000	2100	–
TSS (mg/L)	1000–2000	200–400	600–1900	50–350	100–700	100	–
Color	–	–	Highly colored	Strongly colored	Strongly colored (>14,000 Pt–Co units)	Colorless	Colorless
BOD (mg/L)	2500–3500	100–500	50–120	100–400	50–550	30	–
COD (mg/L)	10,000–20,000	1200–1600	250–400	400–1400	250–8000	250–500	–
Chlorides (mg/L)	–	–	350–700	–	100–500	600–1000	600
Sulphates (mg/L)			100–350		50–300	1000	400

3 Membrane based processes

As already mentioned earlier, traditional treatment techniques suffer from a number of loopholes. The application of membrane based processes in such cases can quite effectively surmount most of these drawbacks. Fig. S1 (Appendix S1) delineates the textile effluent treatment scheme adopted by Arulpuram Common Effluent Treatment Plant, Tirupur, Tamil Nadu, operating under Zero Liquid Discharge conditions. The conceptual flow diagram in Fig. S1 reveals that the treatment facility has been broadly compartmentalized divided into three sections, namely pre-treatment; membrane based recycling unit, chiefly consisting of two-stage reverse osmosis (RO) unit optionally followed by NF and reject management section succeeding the recycling plant (Ranganathan et al., 2007; Singhal and Gupta (RSPCB), 2010). Tables S1 and S2 in the supplementary material (Appendix S1) provide an insight into the characteristics of the textile effluents observed at the inlet and outlet of Arulpuram CETP (Singhal and Gupta (RSPCB), 2010) and Jasol CETP, Rajasthan (Garrett et al., 2012) respectively. A meticulous comparison between the performances of Arulpuram CETP (Table S1) and that of Jasol CETP, Rajasthan (Table S2) indexes the difference in the quality of process water reclaimed in each of these CETPs in terms of the levels of various parameters, such as colour, TDS, TSS, etc.; these observations highlight the fact that the incorporation of membrane technology in the Arulpuram CETP scheme significantly enhanced the quality of the recovered process water, as compared to the engineered Jasol CETP scheme, wherein the TDS and TSS levels marginally increase following secondary treatment through sequencing batch reactor (SBR) and secondary clarifier. Additionally, minimal use of toxic chemicals such as chlorine makes Arulpuram CETP technology innocuous and environmentally benign. Furthermore, sizable recovery of salts and water (approximately 85%) is also achieved in the subsequent RO reject management section. This approach hence satisfies the stringent discharge limits promulgated by environmental regulations and the techno-economic effectiveness incurred by minimization of end of the pipe-treatment.

The choice of suitable membrane technologies depends on the membrane material which is in turn governed by certain vital membrane properties such as it chemical, mechanical and thermal resistance and the membrane susceptibility to fouling; besides, the membrane pore size, which determines the substances that can be effectively retained and membrane shape, which indicates its potential to resist clogging are other important parameters that have to be taken into consideration (Koltuniewicz, and Drioli, 2008; Schäfer et al., 2005). Moreover, management of membrane concentrate streams containing target materials from the feed stream and other auxiliary chemicals added for treatment also poses an arduous challenge to the successful application of membrane based separation processes. Hence, the additional membrane concentrate treatment options have to be explored in terms of cost, energy-intensiveness, efficiency and environmental benignity prior to the discharge of the membrane residuals to the environment (Chelme-Ayala et al., 2009). For instance, direct disposal of the NF and RO concentrates obtained from exhausted dyebath treatment is not an eco-friendly option, given the complex rheology of the concentrates, composed of organic constituents (dyes and additives such as antifoam agents) and inorganic salt components. Reuse of this highly colored waste stream is rendered impossible given the heterogeneity of its constituents, often modified by reactions such as hydrolysis (Van der Bruggen et al., 2003). Bioremediation of the retained dyes through activated sludge system is often not very effective owing to the recacitrancy of the dyes present; in such cases anaerobic degradation used in association with the membrane based processes can be a viable option. Options such as, ozonation and other advanced oxidation techniques, although effective in oxidizing colour, are quite cost-intensive. Bio-decoloring or biosorption coupled with enzyme cataly~~z~~sed degradation are other effective treatment options (Van der Bruggen et al., 2003). However, given the inflexibilities of these biological techniques, membrane distillation (MD) of the NF and RO concentrates followed by incineration of MD concentrates is currently viewed as the most promising scenario, owing to the associated benefit/cost ratio of 3.58 (Vergili et al., 2012). The success of the ZLD approach in such configurations is however governed by the membrane distillation process, which dictates 70–90% of the benefit/cost ratio. The MD distillate is reused in the finishing process, while the value-added energy recovery from the subsequent incineration process offsets the energy consumption in the various other stages of textile effluent treatment (Van der Bruggen et al., 2008).

3.1 Microfiltration

Microfiltration has limited application in textile wastewater treatment because of its close resemblance to conventional crude filtration processes (Mulder, 1996). Microfiltration membranes usually have pore sizes in the range 0.1–~~to~~10 ~~-~~μm; separation through microfiltration is usually effected at a low pressure differential within 2 ~~-~~bar (Dutta, 2007). These features account for its restricted implementation in textile industry. It is mainly used for removal of particles suspension and colloidal dyes from exhausted dye bath and from discarded rinsing bath discharge; microfiltration membranes, however, permit the unconsumed auxiliary chemicals, dissolved organic pollutants and other soluble contaminants to escape with the permeate (Juang et al., 2013; Koltuniewicz, and Drioli, 2008). Hence, microfiltration is seldom used as an unaided independent treatment technique for remediation of complex industrial wastewater such as textile effluents. As such, it is mostly employed as a pre-treatment step in hybrid systems so as to complement other concomitant processes which normally target dyes and other soluble pollutants, but fail to remove the suspended particulate matter present in wastewater. For instance, Ellouze et al. (2012) compared the textile wastewater treatment performances using coagulation/flocculation/nanofiltration, and microfiltration/nanofiltration and observed that for the same volume reduction factor (VRF) of about 8, the nanofiltration permeate flux obtained for feed pre-treated by microfiltration (34 ~~-~~L/h m^2) was relatively higher than the permeate flux of 14 ~~-~~L/h m^2 obtained for feed effluent pre-treated using coagulation/flocculation. This observation was attributed to the considerable retention of color, turbidity, COD and salinity brought about by microfiltration pre-treatment as opposed to the presence of huge quantities of colloidal matter in the feed undergoing coagulation pre-treatment; the colloidal matter present further aggravated the fouling of the nanofiltration membranes. The study, thus established the superiority of microfiltration over coagulation–flocculation as the pre-treatment step prior to nanofiltration, for treating textile wastewaters. Microfiltration can also be used as a post treatment step while treating industrial effluents. For example, Juang et al. (2013) designed a single reactor by combining electrooxidation using BDD/Ti anode and ceramic membrane microfiltration to concurrently remove soluble organic matters and suspended particulate matters, represented by AY-36 azo dye and kaolin respectively, from model textile wastewater. The objective in all these cases was to integrate two suitable treatment methods, which complemented each other by catering to the removal of soluble contaminants on the one hand and targeting the particulate suspensions on the other. Table S3 in the supplementary data (Appendix S1) summarizes the findings of some of the experimental investigations carried out to validate the applicability of microfiltration in textile effluent treatment.

Recently, attempts have been made to fabricate modified microfiltration membranes which can single-handedly attend to industrial textile wastewater treatment. Asymmetric tubular carbon microfiltration membranes were developed by Tahri et al. (2013), using mineral coal powder and thermosetting resin. The as-fabricated membranes were successfully applied to treat industrial textile effluents and achieved satisfactory permeate flux and pollutants removal efficiency, thus bringing about 50% removal of COD, while 30% of the existing salinity was retained; turbidity and colour were almost entirely rejected. Fig. 2 illustrates the experimental apparatus adopted for this research. In another investigation, Daraei et al. (2013) quantified the performance of thin film composite (TFC) membrane, novelly engineered using organoclay/chitosan nanocomposite coated on the commercial polyvinylidene fluoride (PVDF) microfiltration membrane, in retaining methylene blue dye from aqueous solutions. Novel composite membranes were fabricated by Baburaj et al. (2012) from polyamide microfiltration membranes by amalgamating two cationic (poly(ethyleneimine)-PEI, chitosan-CHI) and an anionic (poly (acrylic acid)-PAA) polyelectrolyte through layer-by-layer (LbL) assembly. Two representative textile effluents, methylene blue (MB) and Coomassie brilliant blue (CBB), were employed to evaluate the removal efficiency of the membranes thus developed. The PAA/CHI multilayers achieved a moderate 79.9% CB removal, a satisfactory 87.1% CBB retention and a reasonable 95.35% COD reduction. The membrane fouling tendency was also mitigated by means of this ingeniously engineered arrangement.

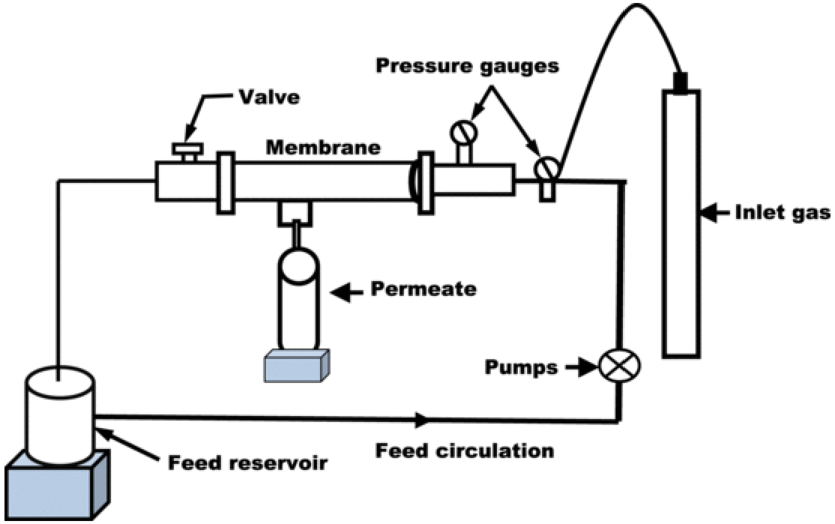

Fig. 2 Schematic of the pilot plant set up for cross-flow mode of microfiltration (temperature: 25 °C, trans-membrane pressure (TMP): range 1–5 bars (Tahri et al., 2013).

3.2 Ultrafiltration

Ultrafiltration is a membrane separation process, mostly used in the separation of macromolecules and colloids from a solution; solutes retained usually have molecular weights of a few thousand Daltons (Dutta, 2007; Mulder, 1996). Although immensely successful in handling contaminants present in the wastewaters discharged from various chemical, pharmaceutical and food industries (Arthanareeswaran et al., 2007; Huang et al., 2012), the ultrafiltration membrane process has limited applications in the textile industry; this is mainly because the molecular weights of the dyes present in the highly colored textile discharge are much lower than the molecular weight cut-off (MWCO) of the ultrafiltration membranes (Ouni and Dhahbi, 2010a; Schäfer et al., 2005). Consequently, dye rejection brought about by ultrafiltration alone usually does not exceed 90% (Schäfer et al., 2005), although higher percentages of dye retention and COD removal have been reported for hydrophobic ultrafiltration membranes, such as, poly-ether-sulfone and poly(vinylidene fluoride) (PVDF) UF membranes (Simonič, 2009; Srivastava et al., 2011). The water reclaimed through ultrafiltration can be reused only in subsidiary processes of the textile industry, such as rinsing and washing, however this recovered water is not qualified for application in primary processes such as dying of fibres, which mandate consistent supply of clean and softened water (Chakraborty, 2010; Koltuniewicz and Drioli, 2008). Ultrafiltration (UF) is usually applied as a pre-treatment step in systems demanding high degree of process stream purification; it is followed by processes such as nanofiltration (NF), or reverse osmosis (RO) stages, which satisfy the demands on process water quality (Barredo-Damas et al., 2010; Simonič, 2009).

Several innovative measures have been examined with an objective to improve the performance exhibited by ultrafiltration technique in treating textile wastewaters.

In an investigation, Marcucci et al. (2001) engineered modules which were innovatively configured to accommodate flat ultrafiltration membranes which operate under vacuum. Koseoglu-Imer (2013) prepared polysulfone (PS) membranes at different evaporation temperatures by phase inversion process and examined the observed concomitant variation in the properties and textile effluents removal efficiencies of the as-fabricated membranes.

Some of the other reported novel mechanisms include polymer/polyelectrolyte enhanced ultrafiltration (PEUF), which involves the complexation of dyes with high molecular weight polymers, followed by ultrafiltration (Mondal et al., 2012; Tan et al., 2006), and micellar enhanced ultrafiltration (MEUF), wherein, surfactants at a concentration exceeding its critical micelle concentration (CMC) is added to a contaminated aqueous solution to form micelles that solubilise organic solutes, which are subsequently separated using ultrafiltration (Zaghbani et al., 2008). For instance, Ounia and Dhahbi (2010b) obtained high 99% and 90% dye rejections, respectively, for Safranin T (ST) and Eriochrome Blue Black R (EBBR) dyes by means of PEUF, using poly (ammonium-acrylate) anionic polymer.

However, conventional polymeric membranes often fail to withstand the testing conditions of the problem environment due to their low resistance to chemicals such as organic solvents, high temperatures or acidic and caustic nature of the solutions (Barredo-Damas et al., 2012). Moreover, progressive membrane fouling and the consequent reduction in permeate flux are common, yet serious handicaps suffered by membrane technology; these drawbacks are inimical to membrane integrity and permeability and eventually lead to considerable decline in the membrane performance. Subsequently significant economic losses are incurred over the entire operational time. This necessitates the introduction of hybrid processes, where ultrafiltration is preceded by a suitable feed pre-treatment process such as, flocculation (Simonič and Lobnik, 2011).

Continuous evolution of the membrane technology has facilitated the development of modified membranes which have been streamlined to exhibit satisfactory resistance to fouling; such modifications have introduced certain novel characteristics in the conventional UF membranes, which have enhanced the competence of ultrafiltration in regard to toxic textile effluents treatment. For example, Srivastava et al. (2011) examined the surface modification of PVDF membranes with varying concentrations of commercially available SAN, an inexpensive copolymer of styrene. The as-fabricated membranes were used to remove colour and COD from aqueous RB5 and CR dye solutions; the synthetic textile wastewater was modelled according to the specifications reported in the collected data, for dye concentrations present in the discharged textile wastewaters from local dyeing companies and common effluent treatment plants (CETPs) in Karur, India. Table 3 lists the various other research works carried out to estimate the competence of ultrafiltration in textile wastewater treatment.

Table 3 Synopsis of the various studies carried out on the applicability of ultrafiltration in textile effluents treatment.

Process description	Membrane specification	Effluents present	Component(s) removed	Permeate flux	Reference
Ultrafiltration (closed-loop recycle)	Membrane: Commercial charged MWCO: 400 Da	Reactive dye (textile yarn and fabric dyeing)	>98% dye rejection >90% electrolyte discharge reduction	Flux: 10 L/m^2 h High water recovery (<90%)	Erswell et al., 1988
Ultrafiltration	Polysulfone Ultrafiltration Membrane MWCO > 700 Da Tubular module	1. Direct Green G, Direct Black Meta, Eriochrome Black T. 2. Raw textile effluents (spent dyebath)	>97% dye (MW > 780) Rejection. Optimum flow velocity: 0.76 m/s COD removal: 75–85% TOC removal: 50–60%	Volume flux: 0.8–1.0 m^3/m^2 d	Majewska-Nowak et al., 1989
Micellar enhanced ultrafiltration (MEUF) (Unstirred batch) Cationic CPC Surfactant.	Organic Polyamide Membrane MWCO: 1000 Da	Aqueous acid dye eosin solution	Retention at 276 kPa: 10% (without surfactant); 73.4% (with surfactant micelles). Optimal surfactant-to dye ratio: 2000 90% surfactant recovery.	Flux: 1. Inversely proportional to CPC loading; 2. Directly proportional to operating pressure.	Purkait et al., 2004a
Micellar enhanced ultrafiltration (MEUF) Surfactants: SDS, CTAB, TX-100. Electrolyte: NaCl	Regenerated cellulose membrane MWCO: 10 kDa (cross-flow ultrafiltration)	Aqueous cationic dye methylene blue (MB) solution	Highest (>97%) retention (anionic surfactant) Optimal pH range: 2–11	Flux inversely proportional to surfactant/electrolyte concentration.	Zaghbani et al., 2007
Micellar enhanced ultrafiltration (MEUF) Cationic CPC Surfactant. MW 358.01	GN polymeric membrane MWCO: 10,000 Da Permeability: 6.28 × 10^{-11} m/Pa s (dead-end ultrafiltration)	Reactive Black 5 (RB5 MW: 991.82) and Reactive Orange 16 (RO16 MW: 617.54)	99.7% RB5 rejection (loading: 1.00 g/L) 99.6% RO16 rejection (loading: 0.05 g/L)	Flux decreases with time and increasing CPC concentration Flux increases with pressure	Ahmad et al., 2006
Tangential ultrafiltration with recycling configuration	$ZnAl_2O_4$–TiO_2 membrane with a zirconia microfiltration interlayer on Moroccan clay macroporous substructure MWCO: 4500 Da Average pore diameter: 5 nm Permeability: 0.26 × 10^{-10} m/Pa s	Salts ((NaCl, $CaCl_2$, Na_2SO_4, $CaSO_4$) cationic dye methylene blue, (MB) (orange acid (OA)	60–93% salt rejection 60–80% (approx.) dye rejection	Flux inversely proportional to concentration	Saffaj et al., 2005
Ultrafiltration	Commercial Multichannel tubular ceramic membrane MWCO: 150 kDa Permeability: 4.04 × 10^{-10} m/Pa s (145.4 L/(m^2 h bar)).	Reactive Black 5 (RB5) dye solution (50–500 mg/L)	Maximum 95.2% dye rejection Optimal transmembrane pressure (TMP): 4 bar Optimal cross-flow velocity (CFV): 2.53 m/s	Maximum flux: 255.86 L/(m^2 h)	Alventosa-deLara et al., 2012

Furthermore, recently, the potential of ceramic membranes as viable replacements of polymer membranes in the UF pre-treatment step was investigated; the preference of ceramic membranes over ordinary polymer membranes was based on their exhibition of comparatively high permeability rates and several other advantageous properties they are endowed with, such as, high mechanical, chemical, and thermal stability (Barredo-Damas et al., 2010). Fig. 3 illustrates the schematic diagram for the experimental set up of the UF pilot plant employed to study the effect of varying cross flow velocities on the performance of three commercial ceramic membranes with molecular weight cut-offs (MWCOs) of 30, 50 and 150 kDa, respectively. The membranes successfully removed 99% of the turbidity while the % retention of the colour constituents ranged between 84 and 98%. The observations established the ceramic UF membranes under scrutiny as feasible pre-treatment alternatives for textile wastewater management. The advantageous aspects of ceramic ultrafiltration membranes have sparked interest in many researchers; as a result, extensive experimentation has been recently carried out to validate the suitability of these membranes in treating recalcitrant effluents, such as textile wastewaters. For instance, a study conducted by Zuriaga-Agustí et al. (2014) examined the separation efficiency and fouling propensity of tubular ceramic ultrafiltration membranes employed to treat simulated textile wastewater characterized by a binary foulant system; the model textile effluent consisted of an azo dye and carboxymethyl cellulose sodium salt (CMC) as the primary colloidal and organic foulants respectively. The membranes exhibited excellent organic matter and dye rejection efficiencies (above 98.5% and 93 % respectively). Additionally, an initial sharp drop in the permeate flux profile was ascribed to rapid blockage of pores.

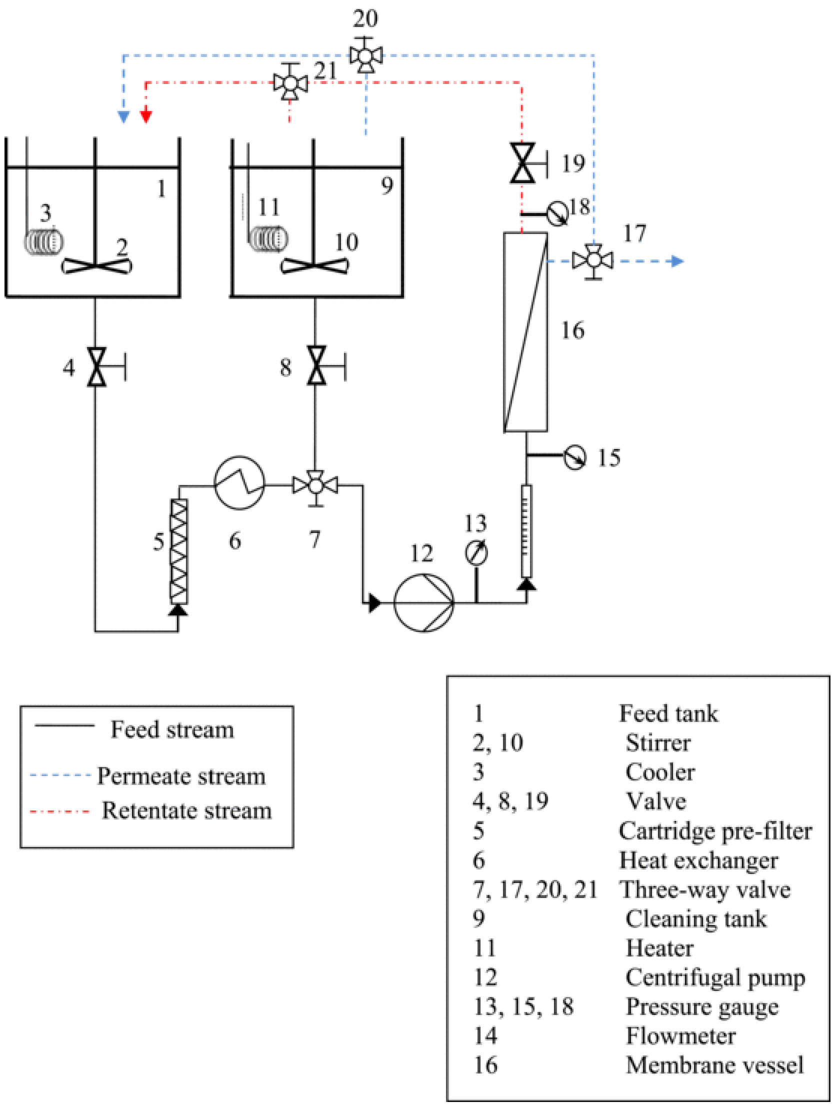

Fig. 3 Schematic representation of the experimental UF pilot plant set up (Barredo-Damas et al., 2010).

3.3 Nanofiltration

Nanofiltration (NF) membrane process is characteristically placed between ultrafiltration and reverse osmosis (Dutta, 2007). Its growing popularity over the years as an effective yet simplified textile effluents treatment technology can be attributed to the several benefits it offers in terms of environmental pollution abatement, rejection, recovery and reuse of textile dyes, divalent salts and other auxiliary chemicals, recovery and reuse of brine. Additionally, the production of quality permeate allows the reuse of treated wastewaters in major processes such as dyeing and finishing (Koltuniewicz, and Drioli, 2008). Nanofiltration operates at a relatively low pressure, which ranges from 500 to 1000 kPa; it enables low retention of monovalent ions, which enhances the scope for low brine rejection and reuse, while permitting almost 100 % rejection of multivalent ions, thus resulting in high solute selectivity. The rejection of species in nanofiltration is governed mostly by steric and charge repulsion. Other advantageous attributes of nanofiltration include its high solvent permeability, retention of dissolved uncharged solutes such as organic molecules, with molecular weight greater than 150 Da ~~Dalton~~, modular construction facilitating scale up, ease of chemical cleaning and the ability of NF membranes to withstand high temperature, up to about 70 °C, which reduces the energy consumed to heat fresh water (Schäfer et al., 2005; Yu et al., 2012).

A number of such investigations have explored the various options available for the enhancement of nanofiltration process performance in the field of textile effluents treatment (Khouni et al., 2011). Yu et al. (2012) examined the performance of submerged nanofiltration of anionic dye aqueous solutions through sodium carboxymethyl cellulose (CMCNa)/polypropylene (PP) thin-film composite hollow fibre membranes; the process was viewed as a beneficial amalgamation of hollow fibre membrane configuration and submerged membrane filtration technology. The hollow fibre membranes are often preferred to flat-sheet membranes, because of their enhanced energy efficiency coupled with high surface to volume ratio; submerged membrane filtration technology, on the other hand, delivers advantages such as relatively lower energy consumption and cleaning requirements than that of tangential filtration mode. It was observed that, the negatively charged CMCNa/PP composite hollow fibre membrane having a~~n~~ MWCO of about 700 Da, was satisfactorily effective in rejection of anionic dyes (Congo red and Methyl blue) from aqueous solution at neutral pH. The percentage dye retention, salt rejection rate and water flux for an aqueous solution containing 2000 mg/L Congo red and 10,000 mg/L NaCl were 99.8%, lower than 2.0%, and 7.0 L/m^2 h bar respectively. Electrostatic repulsion between the dye molecules and the negatively charged surface of the innovatively fabricated membrane was considered as the principal mechanism governing the submerged nanofiltration of saline anionic dye aqueous solutions in the present study.

In another study, Bes-Piá et al. (2010) evaluated the performance of each of the six different spirally wound NF membranes, namely, TFC-SR2, ESNA, NF270, DS-5 DK, DS-5 DL and Duraslick, in treating secondary textile effluents. The behaviour of all the six NF membranes were investigated over a wide range of volume concentration factors (VCF) and the resulting variation in membrane fouling tendency and permeate characteristics were examined. Fig. 4 illustrates the Universidad Politécnica de Valencia designed pilot plant employed to carry out the nanofiltration experiments. The results obtained showed that for all the membranes, normalised flux declined with a corresponding increase in VCF. Nanofiltration membrane charge has significant influence solute rejection and reuse of treated textile discharge. However, most of the commercially available nanofiltration membranes are found to be negatively charged at normal operating conditions with low value pH isoelectric point (Cheng et al., 2012). Very few studies have focused on exploring the performance and the avenues of application that a positively charged nanofiltration membrane can possibly have. However, recently a revival of interest has been witnessed in positively charged nanofiltration membranes and their probable applications. Positively charged nanofiltration membranes hold singular promise in the field of colour removal from dilute wastewater obtained from textile industries, owing to their outstanding hydrophilicity and high multivalent cation retention capacity, which facilitate the recovery of reusable cationic macromolecules and rejection of dyes (Yan et al., 2008). For instance, an unconventional self-assembled positively charged NF membrane (PA6DT-C), originally fabricated in the Swansea laboratory was characterized by Cheng et al. (2012) and its performance was evaluated based on the extent of Methylene Blue (MB) retention and recovery from simulated dye house wastewater. The magnitude of the effective membrane charge density of this novel PA6DT-C membrane was found to be superior to that obtained for two commercially available membranes (Desal-DK and Nanomax-50); also, the membrane flux (∼17 LMH/bar) obtained was relatively higher compared to that observed for the commercial NF membranes (~5 LMH/bar). Excellent performance was exhibited by the NF membrane operating at 5 bar, which achieved a dye rejection of about 98% with a permeate flux of approximately 17 LMH/bar. A diafiltration experiment was carried out to quantify the effectiveness of the membrane in desalting the dye; this evaluation in turn governed the value added recovery and reuse potential of the dye from convention dye house effluents. The results vouched for the successful application of the fabricated PA6DT-C membrane industries such as the textile industrial sector. Table 4 documents the various other investigations carried out by different researchers on the treatability of textile effluents using nanofiltration.

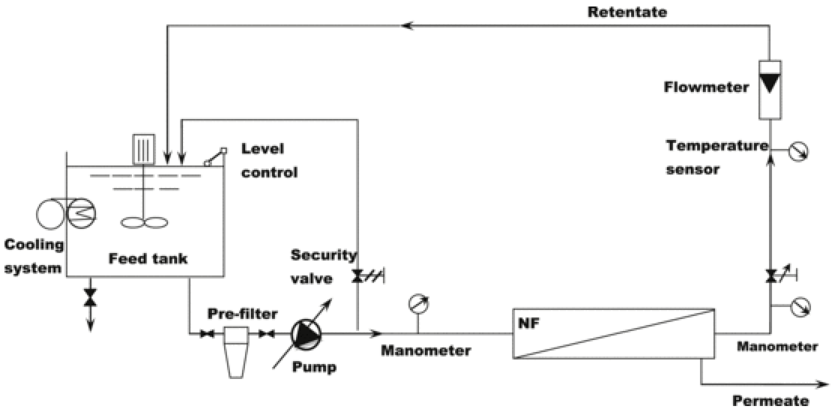

Fig. 4 Experimental pilot plant set-up for NF experiments engineered in the Universidad Politécnica de Valencia (Bes-Piá et al., 2010).

Table 4 Detailed insight into the various studies carried out different researchers on the use of nanofiltration for remediation of textile wastewater.

Process description	Membrane specification	Effluents present	Component(s) removed	Permeate flux	Reference
Ultrafiltration (UF) and nanofiltration (NF)	UF stage: spiral-wound polyethersulfone (PES)	Raw effluents (textile finishing industry)	High color, COD and turbidity removal (NF)	Maximal permeate flux (UF) at pH 11 No significant effect of pH on NF	Alcaina-Miranda et al., 2009

	MWCO: 1 kDa NF stage: commercial spiral-wound (2), NF270 and Duraslick NF		71% salts rejection (NF270) 54% conductivity reduction (Duraslick NF)	permeate flux. Flux (NF270) > (30%) flux (Duraslick NF)	
Textile wastewater reclamation	Three flat sheet NF membranes: Desal-5, NE-70 and TS-40	Textile effluent (dyeing facility)	>99% dye removal Rejection (NE-70) > rejection (Desal-5) Turbidity, hardness, TOC and color removal: <0.2 NTU, 60 mg/L as CaCO3, 10 mg/L and 5 HU, respectively.	Flux (NE-70) twice flux (Desal-5)	Qin et al., 2007
Direct NF and UF/NF	Membranes (NF): NF90, NF200 and NF270 Set up: pilot plant Flat- sheet module	Secondary textile effluent (cotton thread factory). Effluent COD: 200 mg O_2/L TDS: 5000 mg/L	99 % COD reduction. Maximum (95–97%) salt rejection (NF90). Permeate conductivity (NF90) < 500 μS/cm. UF pre-treatment: 40% COD reduction	Flux trend: J (NF270) > J (NF200) > J (NF90). UF pre-treatment: NF permeate flux increase (50%)	Gozálvez-Zafrilla et al., 2008
Fouling tendency examination	Membrane: NF90-2540 Spiral-wound module				
Ultrafiltration (UF) and nanofiltration (NF). Transmembrane pressure range UF: 2–7 bar NF: 4–15 bar.	NF: commercial (NF200 and NF270). UF: polyethersulfone (10 kDa and 1 kDa)	Reactive azo dyes: Everzol Black, Everzol Red, and Everzol Blue. Raw textile effluents (rinsing baths)	COD retentions: 80–100% (UF and NF). Conductivity rates (80%) and Decolorization rates : >90% (NF).	Affected by effluent complexity	Aouni et al., 2012
Reactive dye printing compounds removal by nanofiltration (NF). Cross-flow velocities: 0.4, 0.6 and 0.8 m/s Pressures (2–15 bar).	NFT-50 membrane. Plate and frame module.	Reactive dyes : C.I. Reactive Red 24, C.I. Reactive Orange 12, C.I. Reactive Blue 19, C.I. Reactive Black 5	Dye rejection: 99.4–99.9%. Electrolyte retention: 63–73%. Organics retention: 20–50%	50% Permeate flux decline (adsorption and concentration polarization.	Petrinić et al., 2007
Dye wastewater reuse	Nanofiltration polyamide (PA) composite membranes MWCO: 500 Da.	Separate aqueous solutions of 5 different dyes: Direct Red 75, 80 and 81. Direct Yellow 8 and 27. Model dyeing wastewater: Direct Red 75, PVA, NaCl and Na_2SO_4	Almost 100% dyes rejection. Retention efficiency improvement after coagulation (alum) pre-treatment	20% flux improvement after coagulation (alum) pre-treatment	Mo et al., 2008
1. Comparison between tertiary treatment processes: nanofiltration (NF) and reverse osmosis (RO) 2. UF/NF and UF/RO comparison	1. NF membrane: HL (flat sheet). 2. NF membrane: HL2514TF (spiral-wound) MWCO: 150–300 Da. 3. RO membrane: AG2514TF Configurations: dead end and cross-flow	Raw effluents (denim fabric dyeing factory)	NF permeate quality relatively superior. COD < 90 mg/L. 60% TDS rejection above 9 bar. Permeate hardness: 70 mg/L	11 bar pressure: 9% permeate yield (NF) < 4% permeate yield (RO)	Ben Amar et al., 2009
Nanofiltration using novelly fabricated membranes.	UV-photografting (sodium p-styrene sulfonate monomer on polysulfone UF membrane. MWCO: 1200–1300 Da	Dyes: Acid red 4, Acid orange 10, Direct red 80, Disperse blue 56, Reactive orange 16. Salts: Na_2SO_4, NaCl	97% dye retention (0.4 MPa). Fouling tendency (photografted membrane) < fouling tendency (commercial polyamide membrane Desal SDK	0.23–0.28 m^3/m^2 day (0.4 MPa)	Akbari et al., 2002a
Colour and COD rejection	Spiral wound membranes: MPS 31 (MWCO: not available) NF 45 (MWCO: 200 Da) DK 1073 (MWCO: 300 Da) Cross-flow configuration	Waste waters: 1. Remazol Yellow 3 RS, Remazol BTE Red 3 BS 2. Remazol BTE Blue, RN Special Remazol, BTE Red 3 BS 3. Remazol Black, Remazol BTE Red 3 BS Salts: NaCl, $CaCl_2$, Na_2SO_4	99% color rejection (DK 1073 and NF 45). 87% COD reduction (DK 1073) Fouling tendency (all the test membranes)	30.5–70 L/h m^2	Lopes et al., 2005
Textile dye treatment (polyamide based nanofiltration membrane)	Membrane: Desal 5DK MWCO: 150–300 Da	High molecular weight Direct dyes: direct red 80, direct yellow 8. Anionic dyes: acid orange 10, acid red 4	∼100% dye rejection Rejection (pH 3) Rejection (pH 6) < Rejection (pH 3)	Declining flux profile (fouling)	Akbari et al., 2002b

		Cationic dye: Basic blue 3 Disperse dye: Disperse blue 56 Reactive dye: Reactive orange 16	(cationic dyes)		
Color removal and COD reduction	Membrane: organic membrane (unstirred batch and rectangular cross flow mode) MWCO: 400 Da	Reactive black dye (Cibacron Black B), Reactive red dye (Cibacron Red RB)	94 and 92% dye retentions (reactive black and reactive red dye respectively. 94% COD reduction	Flux profile: (i) rise with increase in trans-membrane pressure (ii) fall with time and increasing feed concentration	Chakraborty et al., 2003
Cross flow nanofiltration	Flat sheet polysulfone based thin film composite (TFC-SR2) nanofilter,	CI reactive black 5 (Bayer, Sydney), Salt: NaCl	Average dye rejection: 98% Average NaCl rejection: <14%	Average flux at 500 kPa: 59.58–78.4% Mean waterflux recovery:99%	Tang and Chen, 2002
Nanofiltration	UV-photografted nanofiltration membranes MWCO increases with increasing hydraulic permeability	Direct red 80 (DR80), disperse blue 56 (DB56), acid red 4 (AR4), reactive orange 16 (RO16) and basic blue 3 (BB3)	Dye retention: >96% at 0.4 MPa	Hydraulic permeability: 0.48–0.56 m^3/m^2 day	Akbari et al., 2006
Nanofiltration	1. Negatively charged polypiperazine amide/poly (phthalazinone ether sulfone ketone) (PIP/PPESK) NF membrane 2. Positively charged quaternized poly (phthalazinone ether sulfone ketone) (QAPPESK) membrane	Synthetic Sulfur Black B dye wastewater	QAPPESK NF membrane better than PIP/PPESK NF membrane 92.3% dye Rejection 10% salts rejection at 60 °C	Flux: 14.5 L/m^2 h (QAPPESK NF) at 60 °C	Han et al., 2009

However, there are still certain unresolved issues which demand feasible solutions before going for large-scale industrial application of nanofiltration. A broad insight was provided by Van der Bruggen et al. (2008) into some of these common impediments, which included membrane fouling due to concentration polarization, degree of separation of target solutes, downstream treatment requirement for concentrates, chemical resistance of membranes and the limited membrane lifetime, insufficient rejection of contaminants; and the necessity for modelling and simulation tools. Measures, such as, monitoring the frequency of membrane cleaning, enhanced selective separation of target solutes by exploiting innovative membrane configurations or by means of judicious selection of membranes and application of new and flexible predictive modelling techniques using equations like Maxwell–Stefan equations were adduced to widen the spectrum of application of nanofiltration, especially in the industrial sector (Van der Bruggen et al., 2008). Additionally, the membrane hydrodynamics can be suitably manoeuvred towards striking a balance between high dye retention and high permeate flux accompanied with low brine rejection (Schäfer et al., 2005).

3.4 Reverse osmosis

Reverse osmosis (RO) is effective in removing macromolecules as well as ions from textile discharge; the treated effluent obtained is usually devoid of colour and has low total salinity (Koltuniewicz and Drioli, 2008). However, the use of dense polymeric membranes and the high osmotic pressure build up due to presence of high salt concentrations considerably delimit the permeate flux, and at times serious fouling takes place, which affects the membrane performance. Hence, in RO, trans-membrane pressures greater than 2000 kPa are necessary in order to maintain reasonable permeate flux, which again deals a severe blow to the process economics (Schäfer et al., 2005).

Liu et al. (2011) conducted a comparative assessment of the effectiveness exhibited by nanofiltration and reverse osmosis in treating biologically remediated textile effluent, based on the evaluation of the permeate quality obtained from each of the processes; the permeate was tested for permeate flux, COD and BOD removal and salinity content. Cross-flow filtration tests of the textile effluent were carried out using BW30 reverse osmosis and NF90 nanofiltration flat-sheet membranes over a wide range of concentration ratios and under different hydrodynamic conditions. The treated process streams, in both the cases, satisfied the reclamation criteria, and reusable water of good quality thus generated with each of the membranes, could hence be recycled to textile processes such as washing and dyeing, thereby saving on water and energy consumption and further costs involved in downstream treatment of water.

Nataraj et al. (2009) further elaborated the performance-wise difference between nanofiltration and reverse osmosis by conducting a comparative study based on the rejection efficiency of the NF and RO modules. Therein the effectiveness of spiral wound NF and RO modules, which constituted a pilot plant, were evaluated in treating a simulated contaminated wastewater mixture in terms of colour and Na_2SO_4 salt rejection over varying feed concentrations and feed pressure with methyl orange (MO) as the model dye compound. Fig. 5 shows the schematic of the pilot plant used along with its NF and RO modules. The rejection of methyl orange obtained through RO (99.99%) was slightly higher than the rejection brought about by NF (99%), while marked decline in permeate flux could possibly be attributed to concentration polarization and membrane fouling. Additionally, for both NF and RO experiments, the TDS removal rates, sodium retention and overall conductivity profiles were similar to that of methyl orange dye.

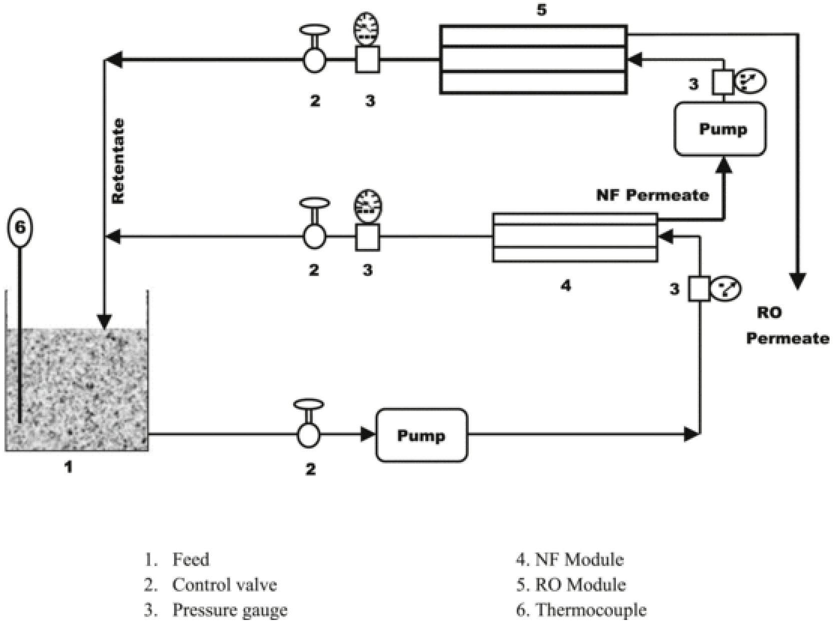

Fig. 5 Schematic diagrams of NF and RO pilot plants (Nataraj et al., 2009).

3.5 Electrodialysis

Electrodialysis is used, rather infrequently, in the textile industries, for abatement of textile wastewater contaminants. Detailed survey of archival literature reveals that there is a dearth of quality studies dealing with the usage of electrodialysis based textile effluent treatment techniques. Electrodialysis is highly functional in removing chlorides and hence, is particularly efficacious in bringing about the legislatively prescribed remediation of wastewaters discharged from Indian textile industries, where the use of bulk quantities of sodium chloride (NaCl) salt is predominant. Moreover, the Electrodialysis with bipolar membranes (EDBM) has recently attracted interest owing to its high energy-efficiency and cost-effectiveness as compared to the more energy-intensive reverse osmosis (RO) processes. Chandramowleeswaran and Palanivelu (2006) investigated the effectiveness exhibited by electrodialysis in affecting the TDS reduction in textile effluents obtained from a common effluent treatment plant (CETP). Treatability study of the textile effluent, thus collected, was conducted in a laboratory scale three compartments electrodialysis system. The experiments were performed in batch, batch recirculation and continuous operative mode under constant current with sodium chloride solution (7500 mg/L) and sodium sulphate solution (5000 mg/L). Operating current density, in the range 3.6–4.8 mA/cm^2 was suggested for handling textile CETP effluent with TDS around 7000 mg/L.

Electrodialysis (ED) can as well be employed to reduce the subsequent volume load on evaporators through concentration of rejects obtained from reverse osmosis (RO) plants. The effectiveness of electrodialysis membrane process in achieving the desired concentration of the RO treated textile discharge was evaluated by Praneeth et al. (2014). The possibility of generating a textile effluent concentrate containing about 6 times the quantity of salts present in the RO reject was explored using a bench-scale system with five membrane cell pairs as schematised in Fig. 6. The limiting current densities, for feed flow rates varying as 18–108 L/h were observed to be in the range 2.15–3.35 A/m^2; the operating cost thus incurred was about one eighth of that resulting from the application of evaporation alone. ED could hence successfully concentrate the RO reject obtained out of textile industrial effluent; the percentage of inorganic constituents in the digest thus obtained, was enhanced from 4.35% to about 24%. ED was found to be especially applicable in energy efficient treatment of RO rejects with low COD concentration; the dilute stream obtained could then be recycled to RO plant for further recovery of reusable process water.

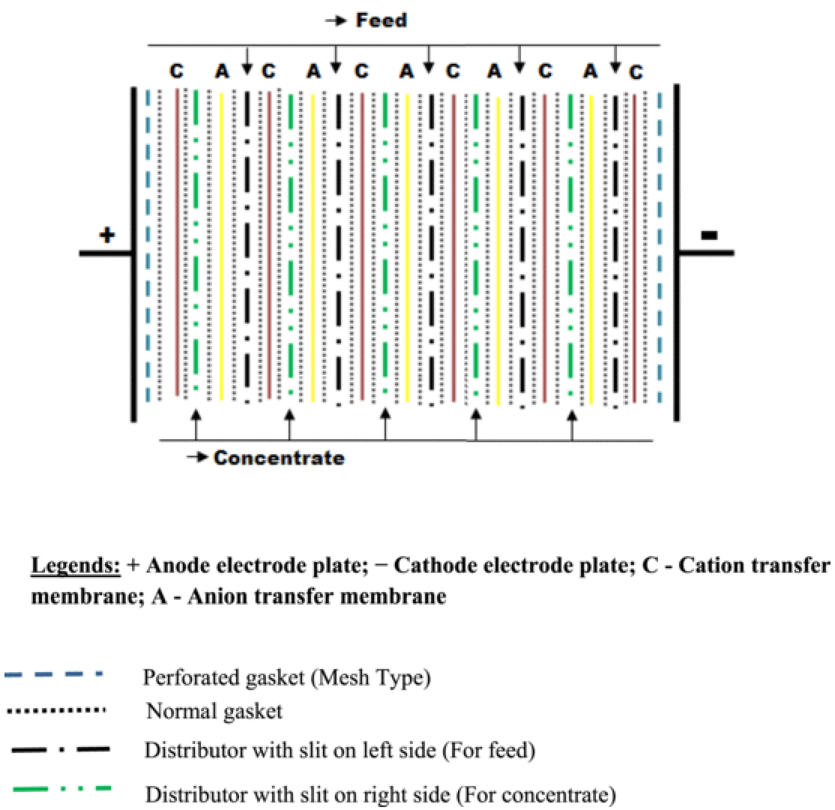

Fig. 6 Engineered ED stack assembly (Praneeth et al., 2014).

3.6 Integrated process

Researchers claim that integrated membrane based processes are highly propitious for the treatment of complex effluents such as textile effluents, which are highly deleterious because of their heterogeneity, and the presence of complex and recalcitrant constituents, such as dyes, salts and auxiliary chemicals (Koltuniewicz, and Drioli, 2008). The consumption of huge volume of materials and high energy in textile industries hence calls for process intensification through the introduction of hybrid treatment techniques; this is done to ensure environmental compliance, and to reduce energy expenditure and materials uptake, thereby abating shock loads on single unit operations (Pophali et al., 2003; Van der Bruggen et al., 2004). The selectivity and effectiveness of these hybrid processes can be suitably enhanced of judicious permutation and combination of treatment techniques which complement each other (Aouni et al., 2009; Ellouze et al., 2012). Literature survey reveals a number of studies conducted to examine the efficiencies of these processes for textile effluent treatment. Many of these investigations focused on the potential of combined physicochemical/biological and membrane separation processes, such as, integrated ozone-BAFs (ozone biological aerated filters) and membrane filtration (RO), for generating recyclable process stream (He et al., 2013). For instance, low-pressure microfiltration with hollow fibre membranes was used by Kertèsz et al. (2014) to separate and recover anatase TiO_2 photocatalyst, which had earlier been employed to degrade organic azo dye, Acid Red 1 photocatalytically. Application of electro-catalytic oxidation equipped with novel tailor fabricated Ti/SnO_2–Sb_2O_3–Y anode prior to nanofiltration of Acid Red 73 (AR 73) wastewater by Xu et al. (2014) significantly improved the subsequent NF process performance by reducing concentration polarization and fouling of membranes. A voltage of 10Vand an operating pressure of 0.6 MPa at low cross-flow velocity was applied to achieve optimal specific energy consumption for the combined process.

In another independent study by Banerjee et al. (2007), the significance of the chronological sequence of the different treatment steps constituting a hybrid treatment scheme was investigated by quantifying the performance of three different effluent treatment schemes in terms of permeate flux and permeate concentration at various operating pressures, feed concentrations and cross-flow velocities; synthetic eosin dye solution was selected as the model textile effluent. Three schemes, namely, Scheme 1 involving advanced oxidation process (AOP) using Fenton's reagent followed by NF, Scheme 2 involving NF followed by AOP and Scheme 3 consisting of two-step NF were selected for examination. The results showed that Scheme 1 was suitable for dye removal for feed concentrations not exceeding 70 mg/L; however, at higher feed concentrations (100–200 mg/L) the reduction of permeate dye concentration below 1 mg/L was rendered difficult. Scheme 3, consisting of two step NF configuration, overcomes this drawback by reducing eosin dye concentration to a level below 1 mg/L at all the feed concentrations (70–200 mg/L); 828 kPa operating pressure and 0.91 m/s cross-flow velocity were selected as optimal process parameters of the scheme. Additionally, supplementary chemicals were not required making Scheme 3 the most feasible and innocuous of all the schemes under scrutiny. Scheme 2 was rejected owing to the significant permeate flux decline observed in the NF unit and more importantly the subsequent unreasonably lengthy AOP process (operating time greater than 3 h) required to bring about permeate eosin concentration reduction below the desirable 1 mg/L level. Table 5 lists the various integrated membrane based processes, involving two or more treatment techniques, which concurrently address the various aspects of textile effluents treatment, including contaminant removal, water reclamation and recovery of unexhausted dyes or auxiliary chemicals. These physicochemical/membrane combinations, however, at times suffer from certain loopholes. Li et al. (2014) reported one such drawback while exploring the feasibility of employing a novel integrated membrane coagulation reactor (IMCR) for the treatment of textile effluents. The increase in coagulant polyaluminum chloride (PACl) dose exacerbated the fouling of membranes in the ultrafiltration unit and the membrane fouling was relatively more pronounced for higher MWCO membrane.

Table 5 Various integrated systems involving membrane based processes for textile effluents treatment.

Integrated systems	Textile wastewater characteristics	Reference
Coagulation/ultrafiltration	Raw textile effluents	Lee et al., 2009
Fenton oxidation/membrane bioreactor (MBR) process	Raw effluent of integrated dyeing wastewater treatment plant (IDWTP) Principal dye: Reactive Blue 4 (RB4)	Feng et al., 2010
Sequencing batch reactor SBR)/nanofiltration (NF)	Synthetic textile effluent: Reactive dyes: Remazol Yellow RR, Remazol Blue RR and Remazol Red RR	Zuriaga-Agustí et al., 2010
Nanofiltration (NF)/anoxic biodegradation	Model post-dyeing textile (knitted cotton fabric) effluent: low temperature reactive dyes Helaktyn Blue F-R (CI Reactive Blue 4), Helaktyn Yellow F-5G (CI Reactive Yellow 1), Helaktyn Red F–5B (CI Reactive Red 2). Real textile wastewater (dyeing and finishing facilities.	Żyłła et al., 2006

Attempts have hence been made to design pragmatic membrane base hybrid treatment systems, dealing exclusively with novel combinations of different membrane based treatment processes, wherein the balance between substance recovery and discharge is essential for achieving higher separation efficiency. For instance, a comparative study was carried out between direct NF and UF/NF hybrid systems by Fersi and Dhahbi (2008) through evaluation of their respective performances in treating textile wastewater. The experimental results highlighted the superiority of integrated UF/NF process over exclusive NF process. Considerable improvement was observed in color retention (> 95%), conductivity and total dissolved salts retentions (80%) and bivalent ions retention (> 95%) values on application of UF/NF combination. Additionally, complete decolourization was achieved even at a relatively low trans-membrane pressure (TMP). Similar studies on the effectiveness of UF as a pre-treatment process for the subsequent nanofiltration (NF) and reverse osmosis (RO) were conducted by Arnal et al. (2008), revealed that incorporation of UF stage prior to the NF stage led to a remarkable decline in the fouling of NF membranes. Nandy et al. (2007) conducted a case study on the employment of ultrafiltration and reverse osmosis in textile effluent treatment. The investigation also elaborated the routing of reverse osmosis (RO) concentrates through evaporator in order to achieve zero liquid effluent discharge. Around 55% of fresh water demand for industry was conserved through the implementation of this treatment scheme. These investigations thus reveal the viability of the integrated treatment schemes which have been ingeniously engineered to address the various issues pertaining to textile wastewater treatment.

4 Modelling and analysis

The discussion on membrane based treatment processes is incomplete without an elaborate perception of the mechanism governing the transport of solute across the membranes and comprehensive modelling of membrane based techniques. Simulation of the performance of various membrane based processes is an indispensable preliminary to the meticulous monitoring of solute transport through membranes (Foley, 2013).

Formulation of a near-accurate predictive model for any membrane based separation technique, hence, has to address the following three basic issues catering to solute transport: transfer of solvents and accompanying solutes (1) within the concentration boundary layer at the phase boundary, (2) through the porous membrane and (3) within the membrane pores (Banerjee and De, 2010a; Dutta, 2007).

The rejection of dyes using PEUF was quantified by Mondal et al. (2012) through a theoretical model which reflected the interaction between dyes and polymers by means of a reaction mechanism obtained from reversible thermodynamics.

The model demonstrating the binding mechanism of the dyes to the polymers was expressed as:

$$[\mathrm{L}]_t = [\mathrm{L}] + \frac{[\mathrm{L}]^2}{K_\mathrm{a}} + \frac{K_1[\mathrm{L}][\mathrm{D}]_t}{1 + K_1[\mathrm{L}]} \tag{1}$$

where K_a symbolises the dissociation constant for conversion of the protonated form of the monomer (LH) to its non-protonated form (L) and K_1 denotes the complex forming constant for the reaction between (L) and independent dye molecules (D) which produces the monomer–dye complex (LD). The influence of the co-ions present on dye retention was also modelled. The calculated values of the various kinetic parameters conformed reasonably to the experimentally determined data, which further validated the reliability of the engineered model equations.

Purkait et al. (2004b) examined the degree of flux decline observed while carrying out micellar enhanced ultrafiltration (MEUF) of an acid dye (eosin red); hexadecyl (cetyl) pyridinium chloride was used as the cationic surfactant in this investigation.

Several efforts have earlier been made to characterize nanofiltration membranes in terms of their pore size distribution effects (Bowen and Welfoot, 2002), or to develop a model which could theoretically assess the performance of membrane nanofiltration in the recovery of valuable products from a process discharge (Oatley et al., 2005). Attempts were further made to improve the permeate flux observed for nanofiltration of dye solutions using turbulent promoters. The performance of the innovatively configured turbulent promoter enhanced nanofiltration of model dye solutions was suitably assayed and successfully simulated using a predictive semi-empirical model (Auddy et al., 2005, 2004).

The resistance-in-series model has been frequently applied in a number of documented modelling-based investigations to evaluate the flux decline occurring in membrane based processes. For instance, Fersi et al. (2009) used the resistance-in-series model to quantify the different filtration resistances governing flux decline in membrane based textile wastewater treatment; the processes included MF, UF and NF, wherein MF and UF preceded the NF process as pre-treatment techniques. The model used was expressed as:

$$R_{\mathrm{tot}} = R_{\mathrm{m}} + R_{\mathrm{cp}} + R_{\mathrm{f}} \tag{2}$$

where resistance, R_m, offered by the membrane material, resistance, R_{cp}, due to concentration polarization, and fouling resistance, R_f were the filtration resistances which contributed primarily to the total filtration resistance R_{tot}. The fouling resistance, R_f was viewed as the summation of reversible fouling resistance R_{rf} and irreversible fouling resistance R_{if}.

The recorded results and the explicit analysis of flux decline using Wiesner and Aptel equations as well as the resistance-in-series model indicated that the flux decline for UF membrane was mainly due to concentration polarization; whereas the decline in flux for NF membrane was attributed principally to fouling.

The salient characteristics solution-diffusion model, film theory and osmotic pressure and the effect of adsorption were combined to seek a mechanistic understanding of the concentration polarization effect which is primarily responsible for fouling of membranes. A steady state mathematical paradigm simulating the nanofiltration of synthetic eosin dye solution was thus developed (Banerjee and De, 2010b).

Banerjee and De (2010a) further amalgamated concentration polarization and pore flow models to quantify the flux decline and solute rejection observed for the nanofiltration of a model textile effluent comprising a two ionic dye (Cibacron Black and Cibacron Red) system and salt (NaCl). The governing factors, namely, diffusion, convection, and electrical migration of the charged species, as well as their interplay were adequately determined.

Lau and Ismail (2009) documented and reviewed models such as, extended Nernst–Plank model (ENP), Spiegler and Kedem model (SP), Teorem–Meyer–Siever (TMS) and the Donnan–Steric pore model (DSPM) used to quantify salt rejection, while unsteady-state mass transfer model was developed to visualize the separation of dye components in an unstirred NF batch.

Kurt et al. (2012) formulated empirical equations using Statsoft STATISTICA 8.0. These equations associated the instantaneous flux with inlet COD, conductivity and color values.

The high COD, conductivity and color retention data obtained revealed the suitability of reverse osmosis (RO-XLE) and nanofiltration (NF-270) membranes for treatment for textile wastewaters.

Srisukphun et al. (2009) designed a mathematical model that quantified the interplay of foulants in textile waste stream, namely, surfactant, reactive dye (anionic dye), and effluent organic matters (EfOMs), and the effect of such interaction on fouling of reverse osmosis (RO) membrane. The permeate flux J could hence be estimated from the following model equation:

$$J = L_0(1 - b\ln(t + t_0) + b\ln(t_0))(\Delta P - \sigma\Delta\pi) \tag{3}$$

where b and t_0 denote respectively the reduction rate of site available for membrane filtration (min^{-1}) and the initial time at which available site reduction commences (min). The notation σ represents the reflection coefficient and $(\Delta P - \sigma\Delta\pi)$ is the effective trans-membrane pressure (TMP) driving force, while L_0 is the initial permeability. $(t + t_0)$ is the filtration time (min).

Besides, batch extraction of model dyes, crystal violet and methylene blue, and their binary mixtures from aqueous solutions using emulsion liquid membrane was mathematically simulated using conventional and modified mass transfer models based on spherical shell approach; the research innovatively explicated the closed form analytical solution for a multi-component system (Agarwal et al., 2010).

Theoretical modelling of membrane based separation processes usually presents several computational challenges. Nevertheless, these models have been quite successful in providing a deeper and a more detailed understanding of the solute transport mechanism across membranes. The parameters, such as permeate flux, membrane fouling tendency and solute retention, can hence be estimated and monitored, albeit with imperfect universality, with the help of these pragmatic simulations (Foley, 2013).

5 Economic evaluation of textile wastewater treatment using membrane based processes

The applicability of any process in the industrial avenue can be ascertained only after analyzing the pragmatism of the process from the economic perspective. Hence, a number of evaluations were conducted by various scientists to verify the economic feasibility of membrane based techniques, so as to ensure the successful employment of these methods in textile industries (He et al., 2011; Ranganathan et al., 2007; Van der Bruggen et al., 2004). The present section illustrates some of these investigations.

A techno-economical evaluation of an ultrafiltration and reverse osmosis based wastewater treatment unit on a pilot scale was carried out by Ciardelli et al. (2000) using secondary effluents from dyeing and finishing plants; the effluents were preliminarily treated by means of activated sludge oxidation. The economic evaluation of ultrafiltration as a textile waste stream pre-treatment technique was also carried out by Simonič (2009) in terms of payback period (t_p), the net present value (NPV), and internal rate of return (IRR) estimation.

Vergili et al. (2012) analysed the effectiveness of the zero liquid discharge (ZLD) method in evaluating the techno-economic aspects of textile wastewater treatment through integrated membrane processes. These hybrid processes comprised various associations of ultrafiltration (UF), loose nanofiltration (NF^1), tight nanofiltration (NF^2) and reverse osmosis (RO). A comparative techno-economic analysis of four different combinations, namely, UF/NF^2 (S (I)), NF^1/NF^2 (S (II)), NF^1/RO (S (III)) and UF/NF^2/RO (S (IV))), was carried out, wherein each scenario was accompanied by a subsequent membrane distillation (MD) process. The evaluated parameters included capital and operating costs, revenues, benefit/cost (B/C) ratios and pay-back times. The third and fourth combinations were not cost-effective, given the decline in the profits achieved by recycling the soda ash and NaCl recovered from the treated wastewater. The unit treatment costs computed for the scenarios S (I), S (II), S (III) and S (IV) were 1.37, 1.38, 2.16 and 2.01 $/m^3 of influent respectively and their corresponding return periods were 0.87, 0.91, 2.07 and 1.51 years, respectively Moreover, the B/C ratios of 3.58 and 3.55 for scenarios S (I) and S (II) respectively, suggested that S (I) and S (II) were the most viable options from the technological and economic point of view. It was thus affirmed that the ZLD approach could most efficiently bring about textile dye bath waste treatment as well as recycling of processed textile waste stream. However, the successful application of this process was delimited the unit incineration cost incurred due to the concentrate which in turn resulted in considerably constrained investment return period; besides the suitability of the approach was primarily affected by the actual volume of processed MD, which contributed significantly (70–90%) to the B/C ratio.

Praneeth et al. (2014) further carried out a comparative economic evaluation of the performance of evaporation alone and that of an integrated process comprising electrodialysis (ED) and evaporation; the performance in each case was scaled in terms of the efficiencies exhibited by the methods while generating textile effluent concentrate through further treatment of RO reject obtained from textile discharge. The estimation, albeit approximate, revealed that the cost incurred to bring about the concentration of RO reject through singular use of evaporation was 3.88 US $ per m^3 of textile RO reject, which was considerably higher than the corresponding operating cost estimated at 0.55 US $/m^3 of textile RO reject for ED-evaporation hybrid technique. The economic analysis hence established the fact that preceding the evaporative treatment of RO reject by electrodialysis was the most economic option, with a relatively short capital investment recovery period of 110 days. A plot illustrating the variation of operating, capital and total costs (*y*~~y~~-axis) against linear feed velocity (*x*~~x~~-axis) further demonstrated a rise in operating cost and a decline in capital cost with a corresponding increase in flow velocity. The optimal linear velocity at a point where the total cost profile hit a minimum level of 0.144 US $/m^3, was reported at 0.008 ~~-~~m/s.

Thus, the explicit application of progressively evolving membrane technology in the arena of textile discharge treatment can successfully bring about process intensification in textile facilities; such an arrangement, in turn proposes to offset the initial capital cost through conservation and extensive reuse of energy, materials and process water. The judicious use of membrane based treatment techniques in textile industries, thereby, infuses an element of sustainability in the process and can be viewed as a promising amalgamation of environmental benignity and cost-effectiveness (Van der Bruggen et al., 2004).

6 Conclusion

The present critical appraisal clearly highlights the fact that the role essayed by these membrane based treatment methods in generating reclaimable textile effluents is quite palpable. The judicious selection of the appropriate membrane based method is, however, influenced by the quality of the treated process stream desired, characteristics of the membrane and the rheological heterogeneity of the effluent at hand, as well as the position of the process in the cost spectrum. For instance, the quality of water recovered through microfiltration or ultrafiltration usually does not satisfy the criteria for reuse in critical processes such as dyeing of fibres; this reclaimed water is mostly reused in rinse vats or as wash water in textile industries. Subsequent NF and~~-~~/or RO processes are therefore necessary for producing premier quality treated effluent that can be directly reused in the primary textile steps such as dying, which demand clean and consistent supply of softened water. Additionally, NF and RO concentrates from membrane based single or hybrid treatment schemes, can be treated further using relatively energy-efficient membrane crystallization units and/or membrane distillation followed by incineration of the MD concentrates so as to bring about successful effectuation of the concept of zero liquid discharge (ZLD). Given the recovery of value-added products from the membrane based processes and the substantial energy yield from the subsequent incineration units, the costs incurred and energy consumed in various prefatory treatment processes are adequately compensated for. The success of the ZLD approach in such configurations is however governed by the membrane distillation process, which dictates 70–90% of the benefit/cost ratio. The present critique also reveals that some of the drawbacks of membrane based techniques, such as membrane fouling, can been adequately resolved through the use of effluent-specific tailor-fabricated membranes or appropriate membrane modules and through punctilious monitoring of pore flow of solutes, concentration polarization and flux decline. The continuously evolving industrial scenario demands further investigations on these membrane based treatment techniques so as to devise ingenious measures to enhance their performance and cost effectiveness. For instance, it is observed that the UF/NF combination is essentially a pragmatic treatment scheme, in terms of performance, membrane fouling control and energy efficiency. Modifications, such as inclusion of PEUF or other conventional treatment techniques, such as coagulation or biodegradation, in such hybrid treatment schemes can be further explored, keeping the mutually conflicting criteria, namely product quality, throughput and cost in perspective. Such treatment schemes can also be considered for scale-up, once the techno-economic feasibility of these hybrid treatment processes is established. Additionally, cumbersome equations encountered while modelling the performance of membrane based techniques, can be solved using advanced simulation tools, such as MATLAB. Nevertheless, despite its imperfections, the genesis of membrane technology can still been regarded as a revolutionary technological breakthrough, and with the future methodological improvements, membrane technology will certainly emerge as the Holy Grail of industrial wastewater treatment.

Appendix A. Supplementary data

Supplementary data associated with this article can be found in the online version, at http://dx.doi.org/10.1016/j.jenvman.2014.08.008.

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Appendix A. Supplementary data

Multimedia Component 1

Highlights

- Role of membrane technology in textile effluent treatment is critically reviewed.
- Parameters and product quality of different membrane techniques are discussed.
- Loopholes in membrane technology are identified.
- Simulations of membrane based textile wastewater treatment methods are summarized.
- Techno-economic evaluation studies on these membrane based processes are appraised.

Queries and Answers

Query: Please confirm that given names and surnames have been identified correctly.

ELSEVIER_YJEMA_4419

Answer: Yes