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Remediation of dyes in textile effluent by membrane based treatment techniques: A critical review

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Abstract: The most important industry implicated in our exposed population is the textile industry arena because of their undeniable contributions to basic human needs satisfaction and to the world economy. These industries are utilizing higher volume of water that finally results in an abundant wastewater generation; these are the major water polluting sources of ecosystem.The present paper provides a review of the management of the membrane technology towards textile effluent treatment. The concept of water reuse is also catching on quickly through membrane based techniques, including the prospect of turning textile sewage into the clarified water. we are using the microfiltration, ultrafiltration, nanofiltration and reverse osmosis.

***Keywords*:** Textile effluents, Membrane technology, Ultrafiltration and nanofiltration.

1. Introduction

Recycled treated or untreated wastewater represents an important health challenge in developing countries due to potential water related microbiological exposure [1].

The textile industry is one of the most polluting industries, due to the large amounts of water and the great quantity of unused dyes and other chemicals released in wastewaters during of the wet processing of textiles [2]. When dyeing/printing textiles, water serves two purposes: First, it ensures the transmission of the colour onto the fibre; second, it washes out excess amounts of dyes from the treated fabrics. Of all dyed/printed textile fibres, cellulose fibres stands out as the most prominent, and more than 50 % of production is dyed/printed with a special class of dyes, the so-called reactive dyes. Over 80,000 tons of reactive dyes are produced and consumed each year, which makes it possible to estimate the total pollution caused by their use [3, 4]. 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, highvolume, and hazardous and toxic wastes [4, 5]. 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 [6– 8]. Besides, these dyes exhibit highly complex structure, high molecular weight and low biodegradability [8–10]. This accounts for its toxic effects on flora and fauna present in the water bodies. Further, these dyes are mutagenic and carcinogenic [8,11].

It is evident that the textile wastewater chemical composition is subject to considerable change due to both the diversity in the textile processes employed and the range of chemicals employed within each industrial category [12]. Wastewater composition is inherently heterogeneous; subsequently, we caution that interpretation of these results should be made within a holistic assessment, including the effects of wastewater composition, dilution, and potential exposure routes within wastewater infrastructure [13].

This has led to increasing environmental concerns; thus, control of water pollution is an important issue. Here we will review some aspects related to the treatment of textile wastewater with special emphasis on membrane processes [14].

The manufacturing industries produce cotton and woolen textiles, carpets, cement, ceramics, and shoes; food processing is economically important and includes sizing where various types of watersoluble polymers, also called textile sizing agents, such as modified starch, polyvinyl alcohol, carboxymethyl cellulose and acrylates, bleaching, mercerizing where sodium hydroxide is added to provide lustrous appearance and strength to cotton, carbonizing (adding acid followed by baking and thereby removing (carbonized) impurities from wool, dyeing and finishing [15].

Consequently wastewater pollutants arise both from raw fabrics and a wide range of additives used to produce the finished product. In textile wastewater treatment, one therefore has to deal with pollutants spanning a wide range including nonbiodegradable highly persistent organics and pesticides used in speciality textiles such as insect-proof fabrics [16]. In particular, the large variability in composition and the presence of potentially reactive components makes textile industry wastewater a particularly diffi cult task for any remediation technology including membrane based methods [17].

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. Membrane filtration in textile wastewater treatment

This method has the ability to clarify, concentrate and, most importantly, to separate dye continuously from effluent [18,19]. It has some special features unrivalled by other methods; resistance to temperature, an adverse chemical environment, and microbial attack. The concentrated residue left after separation poses disposal problems, and high capital cost and the possibility of clogging, and membrane replacement are its disadvantages. This method of filtration is suitable for water recycling within a textile dye plant if the effluent contains low concentration of dyes, but it is unable to reduce the dissolved solid content, which makes water re-use a difcult task [20].

Processes using membranes provide possibilities for separating hydrolysed dyestuffs and auxiliaries, thus simultaneously reducing coloration. Over the last few years, technical and economical improvements have made the treatment of wastewater by membrane systems more and more advantageous over conventional treatment processes [21–23].

The most notable outcome of the membrane treatment is the superior quality of the produced water, which is attained with the addition of fewer chemicals, over conventional water treatment processes. Additionally, membrane plants can be much smaller than conventional wastewater
treatment plants because of the modular confi guration of the membranes, and the possible elimination of other processes (e.g. clarifi cation). Ultrafiltration membranes have been successfully applied in many industries, but it has not been as widely accepted by the textile industry because it does not remove low

molecular weight dyes. There are examples of micelle-enhanced use of UF membranes for dye removal [21,24,25]but in general the rejection range from 30 to 90 % makes the direct use of UF membranes impossible and further fi ltration is required by either RO or NF membranes. In RO, problems with fouling are present which result in low fl uxes due to the dense polymeric membrane used and poor separation.

RO also becomes less effective when osmotic pressure, caused by high salt concentration in the feed wastewater, becomes too high to obtain a reasonable transmembrane permeate fl ux without applying excess transmembrane hydraulic pressure. In this situation NF provides a possible alternative, maintaining high dye rejections albeit with the cost of lower rejection of electrolytes. NF membranes have been used in recovery of salt from used dyeing baths [21,26,27], where the principle is that electrolytes will pass through the membrane, as the dyes will be rejected. However, this principle requires the use of suffi ciently small electrolytes. For example Na_2SO_4 which often is used as electrolyte is rejected by the NF membrane limiting the feasibility of this approach. Also standard electrolytes (such as NaCl) are not high-value products. An NF process for the treatment of mixed waste streams from reactive dyeing process has been developed using this principle where the retentate is further treated in a wet oxidation process. The effective desalination of the retentate by the NF membrane is desired as this diminishes electrolyte corrosion in the wet oxidation. NF based dye retention can be quite effective and retentions up to 99 % with a permeate fl ux of 64 L/m^{2}hr have been obtained [21,28].

RO membranes have also been used in textile wastewater treatment [21,29]. One example of this used a combination of RO membranes designed for brackish and seawater desalination [21,30]. Thus the brackish water RO membrane is used in the fi rst stage and the sea water RO membrane is used in the second stage. In this way the retentate is recycled in the second stage in order to obtain high water recovery and the cross-fl ow velocity is kept high in order to minimise membrane fouling. Using this twostage RO process water recovery is 85 to 95 % with a mean permeate fl ux of 15 L/m^{2}hr.

In a comparative study [21,31] pilot plant tests with UF, NF, and RO treatment of wastewater from washing processes subsequent to reactive dyeing processes are described. UF membrane treatment leaves the permeate stream colorized; whereas NF membrane treatment results in effi- cient permeate decolourization with a fl ux of 70 L/m2hr at 10 bar [21]. The RO membrane both decolourizes and desalinates the waste stream. Interestingly, the reactive dyes retention was somewhat lower than in the NF process despite the use of the denser RO membrane. Severe membrane fouling was observed if the waste stream containing dispersed dyes together with reactive dyes illustrating the important issue of membrane fouling when treating complex wastewaters [21].

In general, pressure-driven membrane processes (PDMPs) include microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO). These filtration processes can be distinguished by the size of the particles or molecules that the corresponding membrane is capable of retaining or passing through [32] This is roughly related to the membrane pore size, which is the main responsible parameter dictating the field of the process applications.

2. 1. Microfiltration

MF is the “loosest” membrane, and it screens particles in the range 0.1-0.5 mm, removing bacteria, protozoan pathogens (Cryptosporidium oocysts, Giardia cysts, and some viruses), silt, colloids, and precipitates. MF is a pressure-driven or vacuum-driven (immersed type) membrane separation process that separates particulate matter from a fluid (water, wastewater textile, or industrial process fluid) by physical straining. MF membranes have demonstrated more than 6-log removal of particles and are credited by regulatory agencies

up to 4-log removal for Giardia and Cryptosporidium. MF membranes remove some viruses and are credited by regulatory agencies up to 0.5-2.5-log removal for viruses, depending on the brand and model [33].

Most MF membranes are manufactured from the polymers that are hydrophilic in nature, such as polyvinylidene fluoride (PVDF), polysulfone (PSF), polytetrafluoroethylene (PTFE), polypropylene (PP), and nylon. The newer version of Teflon membranes were introduced to the market within last few years. Most of the membranes are resistant to oxidants such as chlorine, with the exception of PP. Special agents are often added to the membranes during the fabrication process to reduce fouling of the membranes by dissolved organics [33].

Microfiltration has limited application in textile wastewater treatment because of its close resemblance to conventional crude filtration processes [8,34]. Microfiltration membranes usually have pore sizes in the range 0.1-10 mm; separation through microfiltration is usually effected at a low pressure differential within 2 bar [8,35]. 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 [8,36,37].

Dong et al. (2012) [38] used hydrophilic $Mg(OH)_2$ nanoparticles (NPs) as inorganic fillers to prepare polyvinylidene fluoride PVDF/$Mg(OH)_2$ MF hybrid membranes with enhanced antifouling property. Due to the addition of $Mg(OH)_2$ NPs, a large amount of hydroxyl (OH) groups were formed. This results in an increase of the hydrophilicity of the prepared hybrid membrane, reducing the permeate flux losses caused by membrane biofouling of Escherichia coli and bovine serum albumin (BSA).

Wu, Tang, and Wu [39] developed a novel silica (SiO_2)egraphene oxide (GO) nano hybrid/polysulfone (PS) membrane, which exhibited excellent antifouling ability and improved water permeability, maintaining a high rejection factor to egg albumin. These results were attributed to the specific properties of SiO_2-GO nanohybrid such as the high hydrophilicity and the good dispersion derived from SiO_2 NPs.

2.2. Ultrafiltration

UF is a PDMP placed between MF and NF. However, MF and UF both ivolve similar membrane processes based on the same separation principle, being the difference between both the structure of the membrane, which is asymmetric for UF membrane with a much denser top layer, and consequently a much higher hydrodynamic resistance than MF. UF is commonly used to retain macromolecules and colloids from aqueous solutions and to remove un-dissolved, suspended or emulsified solids from water. Cellulose acetate (CA) and polyelectrolytes are among the first synthetic polymers used for UF membranes. Today, UF membranes are made from a wide variety of chemically and thermally stable synthetic polymers, including PS, PAN, polyethersulfone (PES), polyvinyl chloride (PVC), polycarbonate (PC), aliphatic and aromatic polyamides (PA), polyimides (PI), polyarylsulfone (PAS), Polyetherimide (PEI) and PVDF. 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 [35,40] Although immensely successful in handling contaminants present in the wastewaters discharged from various chemical, pharmaceutical and food industries [41,42], 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 [8,43,44].

The ultrafiltration membrane [45–47] 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 [43,44]. Consequently, dye rejection brought about by ultrafiltration alone usually does not exceed 90% [8, 43], although higher percentages of dye retention and COD removal have been reported for hydrophobic ultrafiltration membranes, such as, poly-ether-sulfone and poly(vinylidenefluoride) (PVDF) UF membranes [8, 48].

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 [8,49] 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 [8,50].

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) [51] engineered modules which were innovatively configured to accommodate flat ultrafiltration membranes which operate under vacuum.

Koseoglu-Imer (2013) [8,52] 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 [8,53,54], 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 [8,24].

For instance, Ounia and Dhahbi (2010) [44] 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.

2.3. Nanofiltration

NF is a PDMP using semipermeable membranes with a molecular weight cut-off (MWCO) in the range of 0.5-2 kDa and pore sizes in the range of 0.5-2.0 nm. The origin of NF dates back to 1970s, when efforts started to be made developing RO membranes with reasonable water permeate fluxes at relatively low pressures [55].
The NF process exhibits separation characteristics between RO and UF. The specific features of NF membranes are mainly the combination of very high rejection factors for multivalent ions (99%) with low to moderate rejection factors for monovalent ions (0-70%), and high rejection factors (90%) for organic compounds with molecular weights above that of the membrane MWCO. The major separation mechanisms of NF involve a steric (i.e., size exclusion) effect and an electrostatic partitioning interaction (i.e., Donnan exclusion) between a given NF membrane and a feed aqueous solution [56].

Nanofiltration (NF) membrane process is characteristically placed between ultrafiltration and reverse osmosis [8,35]. 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 [8, 57].

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, 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[8,43,58]. Recently, there has been a growing interest in developing novel NF hollow fiber membranes because of their flexibility and they are self-supporting and easy to pack in modules with high membrane area per unit module volume [8,59,60].

Bes-Pi_a et al. (2010) [61,62] 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.

2.4. Reverse osmosis RO

The RO process is a reverse of the natural osmosis process, which is the pressure-driven process in which the high total dissolved solids source water is pressurized, and water then passes through the semipermeable membrane. RO membranes have high removal efficiency for monovalent ions such as sodium and chloride and will also remove organic molecules depending on the size and charge of the molecule. Typical RO salt rejection is up to 99.8 % by one single element at standard conditions. However, the overall RO system rejection rate is lower because RO elements are operated in series within the pressure vessels, which results in the increased salt concentration in the feed to each element as water moves along the pressure vessel.
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 [8,57]. 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 (Sch€afer et al., 2005).

Nataraj et al. (2009) [63] 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 Na2SO4 salt rejection over varying feed concentrations and feed pressure with methyl orange (MO) as the model dye compound [8].

3. 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. To date membrane technology for textile wastewater has been based on RO/NF/UF based systems but developments have been hampered by membrane fouling. The membrane technology can still been regarded as a revolutionary technological breakthrough, and with the future methodological improvements.

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