

The Application of Pressure-Driven Ceramic Membrane Technology for the Treatment of Industrial Wastewaters -A Review

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Abstract

This paper presents a review of the previous laboratory analysis and case studies on the application of the pressure-driven ceramic membrane technology for treatment of industrial wastewaters. Ceramic membranes has attracted remarkable interests in recent decades for industrial wastewater treatment because of their superior characteristic such as high fluxes , reliable working lifetime under aggressive operating conditions and ease of cleaning. The literature review revealed that the efficiency of this technology has been proven in a wide variety of wastewaters from different industries and activities including pulp and paper, textile, pharmaceutical, petrochemical, food and mining. However, there are still challenges and questions for this technology that need to be addressed in future researches such as investment cost optimisation by introducing new fabrication technologies, selectivity, permeability and packing densities improvement, fouling minimisation and proposing scale up based on experimental research results.

Keywords: Industrial wastewater, ceramic membrane, Microfiltration, Ultrafiltration, Nanofiltration

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1. Introduction

Generally, Industrial wastewater is an aqueous discharge due to the use of water or cleaning activities in an industrial manufacturing process [1]. Industrial activities generate wastewaters that varies so significantly in pollution characteristics, and each sector of industry produce its own combination of pollutants [2]. These industrial wastewaters may contain heavy metal ions, organic compounds, nutrients, colouring matters, pesticides, endocrine disruptive compounds, and some other toxic materials. As a result, these Industrial effluents should be efficiently treated to protect the environment, aquatic life and humans from intoxication. In addition, due to continuing increase in water shortages and environmental protection concerns, industrial effluent treatment for reuse in the process has been accepted as a sustainable option to address these problems [3].

Ceramic membrane-based treatment system is one of the emerging technologies of treating wastewater that have attracted remarkable interests for the industrial wastewater treatment over the past two decades. The ceramic unit has many benefits over polymeric membranes like high durability, superior chemical, mechanical and thermal stability, bacteria resistance, ability of back flushing and ease of cleaning and sterilization [4-6]. However, there are still challenges for this technology, particularly in optimising capital and fabrication cost, improving selectivity and antifouling properties, enhancing packing densities, and applying experimental research results to large-scale applications [7, 8].

This paper reviews the studies and investigations conducted on the application of pressure-driven ceramic membranes for the treatment of industrial effluents. The paper begins with a brief discussion about ceramic membrane materials and manufacturing followed by a review of the previous laboratory analysis and case studies about the efficiency of this

system in the treatment of wastewaters from different industries. It also covers the challenges and future trends in ceramic membrane technology for industrial wastewaters filtration. Finally, the paper concludes with key findings and recommendations.

2. Ceramic Membrane Technology

2.1 Ceramic Membrane Materials

In terms of separation process, a membrane is described as a selective barrier to separate two phases and it can limit the transport of various elements [9]. There are different categories of membranes based on their materials, such as: 1) polymeric membrane 2) ceramics membrane 3) liquid membrane, and 4) ion exchange membrane [10]. Ceramic membranes for the purpose of wastewater treatment belong to the oxide ceramic membranes which are mainly made of Al, Si, Ti or Zr oxides, and silicon carbide (SiC) and covers the range from Microfiltration (MF) to Nanofiltration(NF). Different oxides have different performance and chemical and hydrothermal stability depending on the operational environment conditions, and therefore can be chosen based on the specific application requirements because each oxide has a different surface charge in solution [11-13].

Desired ceramic membranes for industrial effluent filtration are mainly porous asymmetric structures consisting of a porous support layer , intermediate layer(s) and a thin skin top layer with different densities depending upon the desired molecular weight cut off (MWCO) of the ceramic membranes. All layers can be made of the same material which is called integral or of different materials, which in this case is called composite ceramic membranes and both of them has been used for the filtration of industrial effluents. Figure 1 illustrates the scanning electron microscope(SEM) of the cross-section of a asymmetric composite

ceramic membrane structure made from different materials [14]. Composite ceramic membranes properties and selectivity can be customized by applying different materials in different layers; however, their fabrication process is multi-step and complex [11, 12, 15].

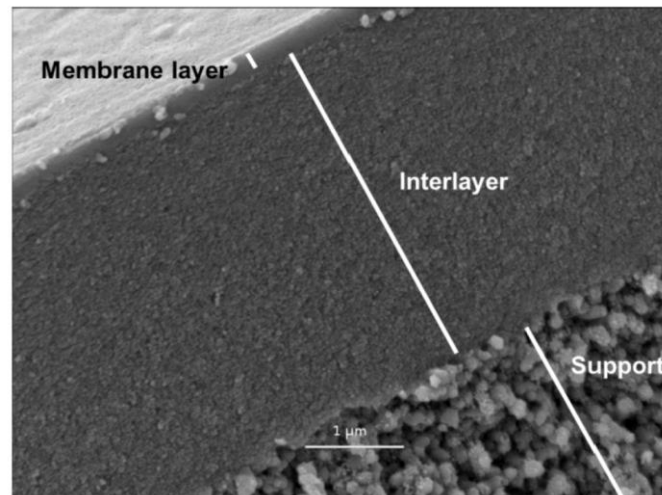


Fig 1. SEM of the cross-section of an asymmetric composite ceramic membrane (reproduced from [14] with permission from the Royal Society of Chemistry)

2.2 Ceramic Membrane Configurations and Their Fabrication Methods

2.2.1 Membrane Configurations

For practical applications, membranes need to be configured into packages that are called membrane modules. They provide a large surface area for an effective feed stream filtration [9]. Ceramic membranes are configured with either a flat geometry and/or cylindrical shapes and of different packaging, volume ratio and materials type to address different operational situations. For the purpose of industrial effluents filtration, cylindrical configuration with single and multi-channel tubes and hollow-fibres are more suitable because of easier sealing of the elements, higher mechanical stability, and better capability to handle higher cross-flow velocities compared to flat geometry [8, 12]. However, because

of ceramic brittleness, optimising the packing densities of ceramic modules in order to reduce the overall footprint of installed treatment unit in operating environment is one of the main concerns of researchers and ceramic membrane manufacturers [12]. Figure 2 shows some photographs of commercially available flat sheet, tubular and hollow-fibre geometries for ceramic membranes.

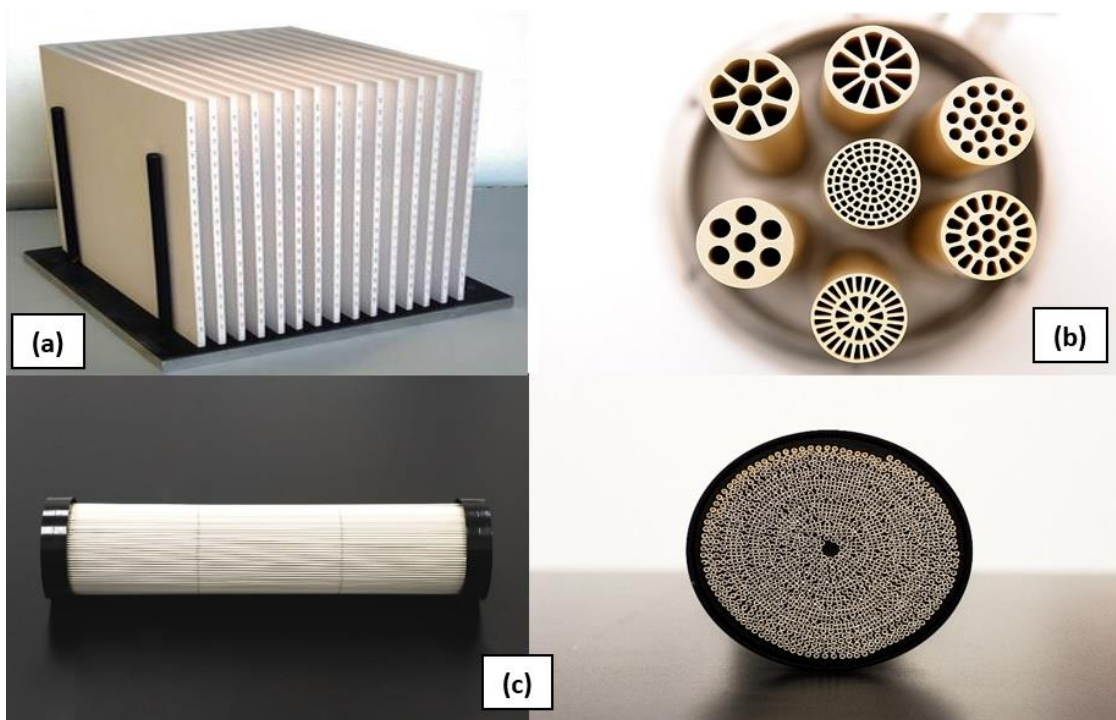


Fig 2. Product photographs of commercial (a) Flat-sheet membrane (reproduced from [16] by permission of © KERAFOLE Keramische Folien GmbH), (b) Tubular ceramic membranes (reproduced from [17] by permission of TAMI industries) (c) Ceramic hollow-fibre Membrane (reproduced from [18] by permission of i2m manufacturing company)

In recent years, commercial ceramic membrane manufacturers have tried to improve the packing densities of ceramic membrane modules. Table 1 lists some current ceramic MF/UF/NF suppliers. Pall® Membralox® developed asymmetric multi-channel tubular alumina MF with pore sizes ranging from 0.1-1.4 μm and zirconia Ultrafiltration (UF) with

pore sizes ranging from 20-100 nm in a unique hexagonal monoliths module to obtain a high packing density up to 240 m²/m³. However, large pressure drop for permeate flow across the monolith is a technical limitation that restricts the diameter of monoliths than can be used [12, 19, 20]. In order to address this problem, Veolia Water Technologies introduced the CeraMem® technology that effectively overcomes the pressure drop problem by mechanical modification to the monoliths. Multiple permeate conduits that conduct permeate through the feed passageways to the permeate collection zone at the end of module were added within the monolith [12, 21]. More recently, TAMI industries Introduced Isoflux™ with flower-like tubular geometries of 8, 23 and 39 channels with membrane filtration area ranging from 0.2-0.5 m². This allows for a stable permeate flux on each point of the membrane independent on the position of measurement which can be beneficial for food processing and bio industry [22]. Hollow-fibre module utilizes tubes with small diameter generally between 2-4 mm thereby compact configuration with highly effective membrane filtration area can be obtained [12]. Furthermore, Fraunhofer IGB introduced a laboratory scale wet-spinning process for the production of porous asymmetric ceramic membranes in capillary module with the outer diameters ranging from 0.5 to 4mm which can improve the packing density of ceramic membranes in hollow-fibre configuration[23]. However, under real operational condition severe fouling and fibre breakage may occur [24]. In recent years, there have been a lot of research on packing density improvement of ceramic membrane modules where good results have been achieved. However, there still an ongoing requirement to design more space-effective modules by fabricating narrower hollow-fibres with an appropriate mechanical strength to reduce the ceramic membrane footprints as much as possible in large scale installation.

138 As mentioned earlier, ceramic membranes find frequent use in industrial wastewaters
139 treatment, and different ceramic membrane materials and modules have been produced
140 worldwide, and many investigations has been conducted to improve the packing densities of
141 ceramic membranes. Yet, commercially available ceramic membranes are still
142 proportionately more difficult to fabricate compared to polymeric membranes and their
143 investment cost is higher than polymeric types. The usage of ceramic membranes has
144 therefore been limited in many real industrial applications and polymeric membranes have
145 dominated the industrial effluents treatment market for decades [\[25\]](#).

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147 Table 1: Some Commercially available ceramic membranes for Industrial effluents filtration

Company	Product	Geometry	Designation	Material of Membrane	Pore size/ MWCO	Available Length(s)-(mm)	Number of Channels	Outer Dia (mm)	Channel Dia(mm)	Ref
TAMI Industries	INSIDE CÉRAM™	Tubular	MF UF Fine UF	-	-	580, 850, 1020, 1178	7,8,11,19,23,25,37, 39,93	25,41	1.6,2.5,3.5, 3.6, 4.6,5.5,6	[26]
	Filtanium™	Tubular	MF UF Fine UF	-	-	580, 1178	8,23,39	25	2.5,3.5,6	
	Isoflux™	Tubular	MF	-	-	1020,1178	8,23,39	25	2.5,3.5,6	
	Eternium™	Tubular	-	-	-	1178	7,8,23	25	3.5,6	
atech Innovations GmbH	atech Ceramic membranes	Tubular	MF & UF	MF: α -Al ₂ O ₃ ,TiO ₂ , ZrO ₂ UF: TiO ₂ ,ZrO ₂ ,Al ₂ O ₃	MF: 1.2,0.8,0.4,0.2,0.1 μ m UF: 0.05 μ m, 150,100,20, 10, 5, 1 KDa	1000,1200 , 1500	1,7,19,37,61,85, 211	10,25.4, 30,41, 52,54	2,2.5,3.3, 3.8,4,6,8,16	[27]
Pall Corporation	Pall® Membralox® IC	Tubular (Hexagonal)	MF&UF	MF: α -Al ₂ O ₃ UF: ZrO ₂	MF:0.8,0.2 μ m UF:100,50,20 nm	1020	48	38,43	4	[28]
Pall Corporation	Pall® Membralox®	Tubular (Hexagonal)	MF&UF	MF: α -Al ₂ O ₃ UF: ZrO ₂	MF:1.4,0.8,0.5, 0.3,0.1 μ m UF:100,50,20 nm	1020	19,37	28,31 38,43	3,4,6	[29]

148

149 Table 1: Continued

Veolia Water Technologies	CeraMem®	Tubular	MF& UF	MF: mixed oxide, α -Al ₂ O ₃ , SiC, TiO ₂ UF: SiC, SiO ₂ , TiO ₂	MF:0.1,0.2,0.5 μ m UF:0.01,0.005 μ m , 50 nm	864	-	142	2,5	[30]
ItN Nanovation AG	CFM Systems®	Flat sheet	MF	α -Al ₂ O ₃	0.2 μ m	L=530 W=6.5 H=110	21	-	3	[31]
Meidensha Corporation	Ceramic flat sheet membrane system	Flat sheet	MF	α -Al ₂ O ₃	0.1 μ m	L=1046 W=12 H=281	-	-	-	[32]
LiqTech International Inc.	CoMem® Conduit	Tubular	-	SiC	-	865	-	146	3	[33]
LiqTech International Inc.	CoMem®	Tubular	-	SiC	-	305,1016,1178	-	25	3	[34]
Inopor®	Ceramic inopor® membrane	Tubular	NF	Support Layer: Al ₂ O ₃ Membrane Layers: TiO ₂ or SiO ₂	MWCO: 750 ,600,450 Da	1200	1,4,7,19,31	10,20, 25,41	3,3.5,6,6.1, 7,15.5	[35]
Cembrane	Cembrane Ceramic membrane	Flat sheet	MF	SiC	0.1 μ m	L=532 W=11 H=150	-	-	-	[36]

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2.2.2 Fabrication Techniques

Figure 3 shows the conventional multi-step process of fabrication of composite ceramic membranes. The choice of method in shaping, heat treatment, and layer deposition processes depends on the application and membrane configuration [11, 12, 15, 37]. One of the main difficulties associated with conventional fabrication processes is the strengthening of ceramic powders and suspensions which requires high sintering temperatures and long sintering times, where consequently grain growth and decomposition of ceramics may occur [38]. In recent years, The combined Phase-inversion and sintering technique has been used as an alternative method for producing wide range of ceramic membranes including flat sheet, hollow-fibre and tubular configurations [39-50]. This method has shown remarkable advantages over conventional methods. Firstly, it is known as one step fabrication process to produce asymmetric ceramic membranes because one heat treatment session is required. Secondly, due to formation of finger-like micro-channels associated with this technique, significant reduction in mass transfer resistance for permeation flux can be observed during operations [12, 48]. This technique consists of preparation of a suspension containing ceramic particles, organic solvent, polymer binder and water. Due to solvent/non-solvent exchange induced phase inversion process, solidification of ceramic suspension occurs and ceramic particles are immobilized by spinning or casting depends on required geometry. Finally, the membrane precursors formed with this method go through a one-step heat treatment to remove all organics and strengthen mechanical properties [12, 40, 50]. Current research has made remarkable progress in determining the effects of different parameters on final selectivity and mechanical strength of ceramic membranes produced by this method. Kingsbury et al. (2009), Tan et al. (2001) & Wei et al. (2008) found that the pore size and selectivity of the final membrane is highly affected by size distribution

of ceramic particles in the suspension. They succeeded to achieve various MWCs in the range of hollow-fibre MF and UF by applying different alumina and zirconia particle sizes in the suspension. In addition, they realized that changes in ceramic to polymer binder ratio have different effects on the mechanical strength of the membrane [40, 44, 45]. Kingsbury et al. (2009) & Tan et al. (2011) studied the morphology of different asymmetric alumina hollow-fibres made by combined phase-inversion and sintering. Two basic sub-structures including finger-like and sponge-like have been observed within the membrane cross section. They found that by changing spinning process parameters including viscosity of the spinning suspension, bore fluid composition and flow rate, hollow-fibre morphology can be varied remarkably [40, 51]. On the other words, variation in spinning parameters leads to different dimensions of the finger-like and sponge-like sub-structures, consequently different morphologies will be generated.

As a result, combined phase-inversion and sintering technique has a great potential to produce ceramic membranes for industrial wastewater treatment applications. However, one of the main drawbacks of this technique is high sintering temperature which has an adverse effect on surface porosity and mechanical strength of resultant ceramic membranes. Advanced sintering methods such as Controlled sintering process by using polyethersulfone (PESf) as a pore structure stabilizer [46], microwave sintering [52-55], spark plasma sintering [56-59] and high frequency induction heat sintering [60-62] have been introduced for inhibiting grain growth during ceramic powder consolidation and for reducing the sintering process temperature and time. Despite remarkable advantages of these advanced sintering methods over conventional sintering techniques such as large energy saving, better grain distribution and enhanced mechanical properties of produced ceramics [52, 53, 55], controlling grain growth during sintering process is still a major issue,

which makes nanostructured ceramics hard to fabricate [38]. Therefore, further investigations are still needed to introduce non-complex and economical fabrication methods with lower temperatures of sintering and shorter processing times to minimise the problem of grain growth, and makes the production of nanoceramics with lowest grain size possible.

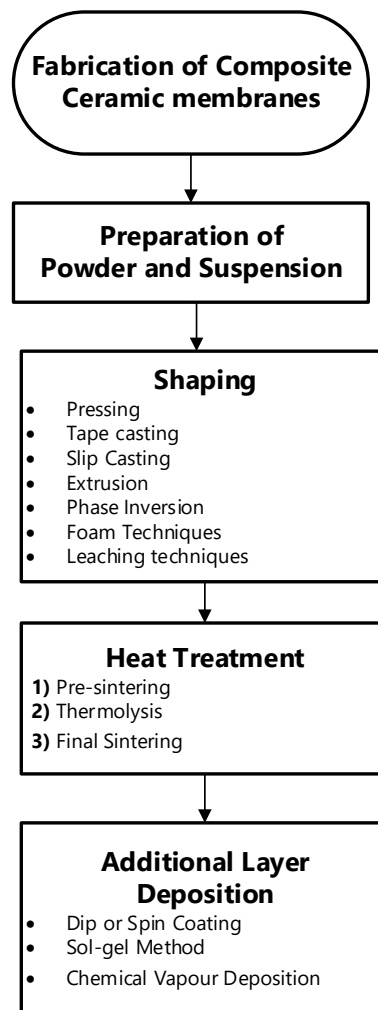


Fig 3. Conventional fabrication of composite ceramic membranes(Adapted from [12])

3. Application for the Treatment of Industrial Wastewaters

One of the most serious environmental issues in the world is the existence of harmful and toxic pollutants in industrial wastewaters. The major industrial categories are mining, food, pulp and paper, textile, petrochemicals and pharmaceuticals. All of the mentioned industries produce wastewaters that have adverse impacts on each components of the environment such as water bodies, soil, air, human and the ecosystem [63]. Therefore, having a reliable wastewater treatment technology is important as it helps in reducing harmful impacts associated with industrial wastewaters. Due to economic and technical limitations of conventional wastewater treatment methodologies, many industries turn to membrane technologies to perform more reliable wastewater treatment operations. Among the various membrane materials, ceramic membranes have been gaining attentions for industrial wastewaters treatment because of their robustness and lower operational costs compared to polymeric membranes. However, despite the advances achieved in this technology, the potential of ceramic membranes for industrial wastewater treatment has not yet been fully realised. Ceramic membrane systems still require improvement in terms of investment cost, packing densities, selectivity and antifouling properties to satisfy future harsh operating conditions. It is crucial to review the investigations of the application of this technology to treat different industrial effluents to give a clear vision for the future [4]. It is therefore, the objective of this section to review some case studies and laboratory analysis of the application of ceramic membranes in some major industrial sections such as textile, pulp and paper, petrochemicals, mining, food, and pharmaceuticals to realize advantages , challenges, and prospects in ceramic membrane technology for industrial wastewaters filtration .

3.1. Pulp and Paper Industry

Pulp and paper industry depends heavily on a massive amount of water and its quality in the various stages of manufacturing processes. The manufacturing process mainly includes wood pulping and production of paper and generates wastewaters containing huge amount of pollutants depending upon the stage of the process. Generally, these pollutants are characterized by chemical oxygen demand (COD), biochemical oxygen demand (BOD), suspended solids (SS), level of toxicity, and colour [64]. The amount of pollutants discharge can also change significantly even at the same manufacturing process due to the use of various chemicals and seasonal variations. To protect human health and environment, many government agencies are forcing the paper industry around the world to treat wastewaters to comply with the environmental guidelines and standards before discharge. In addition, due to shortage of freshwater sources in some countries and increased legislation demands [65-67], the pulp and paper industry use advanced water treatment systems for effluent treatment and reuse it in manufacturing process. In the last two decades, membrane separation technologies have attracted more attention as an unconventional method for the treatment of the paper mill effluent. Successful demonstrations of membrane technology in treatment of wastewaters generated in paper manufacturing processes have been reported by researchers and paper manufacturing companies.

Ceramic membranes have been proposed for the treatment of pulp mill wastewater because of their remarkable chemical, thermal and mechanical stability under harsh operational conditions. Compared to polymeric MF, UF, and NF membranes [65-71], ceramic membranes can be cleaned with variety of harsh cleaning agents when fouling or scaling occurs, and solid performance over longer periods of operations can be guaranteed

because of the nature of ceramics that have a combination of stronger bonds which is partially ionic and partially covalent [12, 72]. Several studies have been conducted to evaluate the performance of various ceramic membranes to treat pulp and paper industry wastewaters. Table 2 illustrates an overview of some investigations evaluating the application of ceramic membranes to treat effluents from pulp and paper mills. As shown in Table 2 , mainly α -Al₂O₃ ceramic membranes with a selective separation layer of TiO₂ or ZrO₂ which are easily available in market with different pore sizes and MWCOs under various operational parameters have been applied in the experiments. Nataraj et al.(2007) developed a pilot plant which was a combination of α -Alumina tubular ceramic MF followed by Electrodialysis (ED) technology for the first time to treat real effluent samples from a paper mill with a COD concentration of 390 mg/L [73]. Their finding showed that ceramic MF plus ED hybrid process was capable to recover 80 percent of wastewater, while the remaining retentate could be used as a biomass. Their proposed plant was found to be more beneficial because the ceramic MF pretreatment could tolerate higher temperature of discharged wastewaters around 60 °C. Fouling during filtration was not severe and was reversible by membrane cleaning using hydrochloric acid and sodium bisulfate. Ebrahimi et al.(2015) employed two different multi-stage ceramic membrane process including hybrid Al₂O₃ MF followed by Al₂O₃/TiO₂ UF and Al₂O₃/TiO₂ UF followed by TiO₂ NF to treat alkaline bleaching effluent from sulfite pulp production with 10600 mg/L COD concentration [74]. Experiments were focused on investigating the suitability of ceramic tubular membrane systems as another possibility to conventional wastewaters treatment methods applied in paper mills. A comparison between two ceramic membrane processes showed that in terms of separation efficiency, the two-stage process employing MF plus UF was the best option for an efficient treatment of the bleaching effluent. More than 35 % of COD and 70% of

297 remaining lignin level was reduced by applying the mentioned configuration. Also,
298 approximately 186 and 140 L/m²h of permeate flux were observed for ceramic MF and UF
299 respectively. Membrane fouling is reduced significantly by applying ceramic MF prior to the
300 UF stage. On the other hand, flux decline over time was observed during the UF/NF process.
301 It was concluded that by applying hybrid ceramic MF/UF configuration, the volume of
302 untreated bleaching effluent discharged from paper mills, would remarkably be reduced
303 when applied to large-scale operations.

304 The other important application of ceramic membrane processes is recovering valuable
305 materials from pulp and paper wastewaters. Researchers [75-84] employed various MF/UF/
306 NF ceramic membranes to treat and extract the lignin from black liquor generated in wood
307 pulping process (Table 2). Black liquor is one of the waste streams in the paper production
308 and is generally characterized by BOD, COD, dissolved inorganic compounds, lignin
309 derivatives and bark particles. Lignin separated from black liquor during ceramic membrane
310 filtration can be used as biofuel, dispersant, blinder, emulsifier and precursor for carbon
311 fibres [75]. According to studies [75-85], by changing the MWCO of ceramic membranes, the
312 molecular mass of lignin can be controlled during fractionation. Jönsson et al.(2008)
313 employed hybrid ceramic UF membrane followed by polymeric NF membrane to compare
314 lignin fractionation efficiency of the combined system with the direct polymeric NF [78].
315 Based on their results, a higher purity of lignin was achieved by applying ceramic UF
316 pretreatment before polymeric NF stage. Furthermore, Žabková et al.(2007) used Al₂O₃-TiO₂
317 tubular UF ceramic membranes with 1,5, 15 KDa MWCOs to efficiently recover vanillin from
318 lignin/vanillin mixture of various concentrations [86].

319 The application of ceramic MF membrane for the pretreatment of paper mill effluents prior
320 to RO has been investigated by Pizzichini et al.(2005) [87]. The wastewater contains 1089

ppm of COD, 330ppm of SS and 435ppm total organic carbon (TOC). Their results showed that applying ZrO₂ ceramic MF with MWCO of 0.14 μ as a pretreatment step prior to RO can guarantee higher steady state flux rate around 150-200 L/m² h and less fouling indexes compared to all other polymeric MF and UF membranes investigated in their experiments. Integrating ceramic MF pretreatment followed by RO post treatment allowed the reuse of more than 80% of effluents as pure water, and it demonstrated the good potential to develop a large-scale industrial process appropriate for providing a large portion of water recovery, with an excellent chemical composition.

According to investigations mentioned in Table 2, there was no evidence of irreversible fouling observed when various cleaning methods such as washing and rinsing with tap water, permeate, deionized water, Alkaline and acid cleaning (NaOH and HCl solutions) were used for membrane cleaning. Approximately between 80-98 percent of pure water flux is restored by proposed cleaning method for MWCOs ranging from 1kDa to 20 KDa whereas for polymeric membranes , only up to 80% of pure water flux can be restored by a combination of rinsing and chemical cleaning [[84](#), [88](#)].

Based on the surveys presented in Table 2, applying ceramic MF/UF pretreatment can guarantee reasonable turbidity, COD, BOD and SS reduction in paper mill effluents. Using ceramic MF or UF pretreatment is beneficial in terms of obtaining stable operations and producing feed water of satisfactory quality for the NF and RO post treatments. Ceramic membranes are appropriate in dealing with aggressive and harsh environment due to high mechanical and chemical stability and higher durability compared to polymeric membranes. Recently, TAMI industries, one of the most prominent companies in the field of ceramic membrane manufacturing, introduced INSIDE CeRAM™ ranging from MF to fine UF with

non-circular multi-channel tubular geometries and with membrane filtration area varying from 0.16-0.6 m² for the treatment of coating effluent in the paper industry [89]. However, the high capital cost and lower packing densities due to fragility of ceramics compared to polymeric membranes are the key challenges for many large-scale pulp and paper operations. Many pulp and paper plants around the world suffer from lack of space that may restrict the ceramic membrane unit installation. The capital cost of the ceramic membranes varies with the MWCO of the membrane. Typically, cost increases with the increase of membrane selectivity in terms of MWCO. Arkell and co-workers reported that the cost of TiO₂ NF ceramic membranes with 1kDa MWCO is about 2000 €/m² [75]. The cost decreased to 470 €/m² for Al₂O₃-TiO₂ Ceramic UF with 20kDa MWCO [75]. Moreover, the cost of UF ceramic membranes is about 33 times more than the polyamide RO membrane but their lifetime is longer. As an example, in one of the industrial installation of ceramic membranes unit in Japan, there was no report on the replacement of ceramic membrane unit after 16 years of successful operations [90]. Due to ongoing importance of ceramic membrane process for wastewater treatment purposes, the future researches may focus on developing wider range of cheaper inorganic membranes by introducing new fabrication technologies and new modules design to offer higher filtration capacities for employing in paper plants. Also, from technical and economical point of view, a feasibility study on the integration of ceramic membrane system with other physico-chemical treatment methods seems necessary [70].

Table 2: an overview of some investigations evaluating the application of ceramic membranes to treat effluents from pulp and paper mills.

Operational Parameters	Feed Source& parameters	Membrane characteristic	MWCO/Pore size	Flux	Rejection efficiency	Cost	Ref.
Pressure:1-5 bar Temp: 25°C Feed circulation velocities: 0.7, 1.3 and 1.8 m/s	First caustic extraction stage of a Kraft pulping Mill	Tubular Ceramic UF with an active layer of ZrO ₂	MWCO of 10000 Da	35-87 kg/m ² h	Colour separation:59-74% TOC:49-53% TS:17%	-	[91]
Pressure:1-5 Bar Temp: 25°C Feed circulation velocities: 0.7, 1.3 and 1.8 m/s	First caustic extraction stage of a Kraft pulping mill	Tubular ceramic MF plus UF with an active layer of ZrO ₂	MF: Avg. pore Dia of 0.14 µ UF: MWCO of 10KDa	53-78 kg/ m ² h	Colour separation:39-52% TOC:28-36% TS:10-14%	-	[91]
Pressure :4 bar Temp:25°C and 60 °C	Real effluent samples from West Coast paper mills, India Conductivity (mS/cm) :10.78 TDS (mg/L): 6046, Lignin (mg/L): 50 DOM (mg/L): 9.166, COD (mg/L):390 BOD (mg/L) :35	α-alumina Tubular Ceramic MF +ED	MF: 20 kDa MWCO and pore size of 1.5µm	113 L/ m ² h at 25°C 121 L/ m ² h at 60°C	Conductivity (mS/cm): 0.5 TDS (mg/L) :250, Lignin (mg/L) :5 DOM (mg/L) –, COD (mg/L) – BOD (mg/L) –	-	[73]
Transmembrane pressure(TMP):2-20 bar, Temp:90 °C CFV: 4–2 m/s	Untreated Kraft black liquor TDS (g/L) :183 ± 2.7 Total hemicelluloses (g /L) :3.56 ± 0.12, Total lignin (g/L): 63.8 ± 1.3	TiO ₂ Ceramic NF	MWCO:1KDa	Avg.: 159 L/ m ² h	Retention of lignin and hemicelluloses :80 %	Membrane Price:1000€/m ²	[75]
TMP :2 bar, Temp 90°C CFV of 5 m/s	Untreated Kraft black liquor TDS (g/L): 183 ± 2.7 Total hemicelluloses (g/L) :3.56 ± 0.12, Total lignin (g/L): 63.8 ± 1.3	Al ₂ O ₃ –TiO ₂ Ceramic UF	MWCO:20K Da	-	TDS (g/L): 176 ± 6.3 Total hemicelluloses (g/L) :2.12 ± 0.06 Total lignin (g/L) 57.6 ± 2.4	Membrane Price: 470 €/m ²	[75]
TMP:3,5, 7 Bar Temp:30±2°C	Untreated Kraft black liquor from wood pulping	α-alumina tubular ceramic UF/NF membranes with an inner layer of either TiO ₂ or ZrO ₂	MWCO: 1 KDa, 5KDa, 15 KDa	52 L/ m ² h for 15 KDa 12-25 L/ m ² h for 1KDa 30-75 L/ m ² h for 5KDa	Retention of organics between 60-70%	-	[76]
Pressure:0.1-0.6 MPa Temp: 25-80°C Flow speed: 1m/s	Untreated Kraft black liquor Feed Lignin concentration (g/L) : 48.8-78.6	ZrO ₂ ceramic UF/NF	MWCO: 1 KDa, 5KDa, 15 KDa	TMP MWCO, and temp all affect the flux	Permeate lignin concentration(g/L) : 15.9-33.8	-	[77]
UF : TMP:100kPa,Temp: 90 °C CFV:5m/s NF: TMP:2.5MPa,Temp: 60 °C CFV:4m/s	Hardwood Black liquor TDS: 17 % wt. Ash(g/L):0.44 Lignin(g/L):59 , Hemicelluloses(g/L) :609	Hybrid Ceramic UF and polymeric NF	MWCO of UF: 15kDa MWCO of NF: 1 KDa	CFV, TMP temp all affect the flux	UF permeate: TDS:16 %, Ash(g/L):0.47, Lignin(g/L):54 Hemicelluloses(g/L):2.5 NF permeate: TDS:11%, Ash(g/L):0.61 Lignin(g/L):13, Hemicellulose(g/L):0.3	Production cost of €33 per tonne of lignin-UF and NF investment costs:(3300 and 2000 €/m ²) respectively	[78]
TMP:200kPa Temp:90°C	Black liquor TDS: 22% wt. Lignin(g/L):62 Ash content :43%	Al ₂ O ₃ -TiO ₂ UF ceramic membrane	MWCO of UF: 15kDa	Avg. flux rate between 110-160 l/ m ² h	Approx. 35% lignin retention	Avg. 20 € per MWh of calorific value of the lignin fuel	[79]
Temp:150 °C	Kraft black liquor Lignin(g/L): 55.9-61.9	ceramic UF membrane	MWCO of UF: 5 and 15kDa	-	Approx. 70% lignin retention	-	[80]
MF: TMP:2-4 bars,Temp:120°C CFV:4 m/s UF: TMP: 2 bars,Temp:70-85°C CFV:5-8 m/s	Black Liquor Dry solids content:40.6-40.8% Total carbohydrate content : (g/kg DS):20-38 , lignin (g/kg DS):337-389	MF/UF ceramic Membranes	MWCO:300k Da-15kDa MF pore size:0.2 µm	-	Dry Solids content:27.6-37.9 % Total carbohydrate content(g/kgDS):9-23, lignin(g/kg DS):242-399	-	[82]

369

370 Table 2: Continued

For 5 KDa TMP:400 kPa,Temp:90°C CV:3.6 m/s For 15 KDa TMP:100k Pa, Temp:90°C CV:4.5m/s	Kraft black liquor Total dry substance(TDS):16 wt. % Lignin content (g/L) :56 Inorganics (g/L) :37	UF ceramic membrane	MWCO of UF: 5 and 15kDa	flux rate 451 L/ m ² h for 5kDa and 951 L/ m ² h for 15 kDa	66% lignin recovery for 5 KDa and 28% for 15 kDa	-	[81]
TMP:155 kPa pH:8.5-12.5	Lignin/Vanillin mixture lignin/vanillin mixture with different concentrations (g/L): 60/6-60/5-5/0.5-20/2	Al ₂ O ₃ -TiO ₂ Tubular UF ceramic membrane	MWCO of UF: 1,5 and 15kDa	14 L/ m ² h for 5kDa and 4 L/ m ² h for 1kDa for 60/6 mix	Best rejection of lignin observed by using 60/6 g/L lignin/vanillin mixture and by employing 1 KDa MWCO at pH :12.5	-	[86]
-	Black liquor TDS (Total dissolved solids):10.3 % Inorganics:77% Organics:23% Lignin:17%	TiO ₂ Tubular UF ceramic membrane	MWCO of UF: 5,10 and 15kDa	-	Permeate parameters: TDS:6.71-9.48 % Inorganics:82.6-92% Organics:8.05-17.4% Lignin:16.9-81.5%	-	[83]
TMP:200kPa Temp:32±2 ,63±3°C CFV:1.2 m/s	Black liquor TS(g/L):82-92 COD(g/L):75-89 Lignin(g/L):26	α- Al ₂ O ₃ tubular ceramic MF membrane	Pore size:0.2µm- 0.8µm	Avg. flux 200 and 400 L/ m ² h at Temp 32±2°C and 63±3°C respectively	Approx. 80 % of lignin retention achieved	-	[84]
TMP: 1 or 2 bar Temp:60 °C, CFV :4–5.6 m/s	Alkaline bleaching effluent COD(mg/L): 10,400 TOC(mg/L):4000 Na(mg/L): 2430	Hybrid Al ₂ O ₃ MF and Al ₂ O ₃ /TiO ₂ UF	MF: 0.1 µm, 0.14 µm, 0.2 µm UF: 5 kDa, 20 kDa, 0.05 µm	For MF and UF 186 and 140 L/ m ² h respectively	Using 0.1µm MF and 20-kDa UF COD removal 35%-45% Lignin removal:60-73%	-	[74]
TMP: 2 bar Temp:60 °C, a l CFV :0.3 m/s	Alkaline bleaching effluent COD(mg/): 10,400 TOC(mg/L):4000 Na (mg/L): 2430	Hybrid Al ₂ O ₃ /TiO ₂ UF And TiO ₂ NF	UF: 5 kDa, 20 kDa, 0.05 µm NF:1 KDa	UF Flux between 36.2 to 5.1 L/ m ² h after 6 hours	Using UF, 20 kDa and NF, 1 kDa COD removal 35%-40% Lignin removal:45-66%	-	[74]
MF: TMP:11 bar, Temp:30°C Feed flow: 4200 L/h	Raw Paper mill effluent SS (ppm):345,COD(ppm):1089 TOC(ppm): 435 Conductivity(µS/cm):2940	Tubular ZrO ₂ Ceramic MF	MF MWCO:0.14 µm	150-200 L/m ² h	MF permeate: SS (ppm):0 COD (ppm O ₂):891, TOC(ppm):361.4 Conductivity(µS/cm):2760	-	[87]

371

3.2. Textile Industry

The textile processing is one of the oldest and largest consumers of water. Textile production is complex and involves spinning, weaving, dyeing, printing, finishing and garments manufacturing. In almost all these stages of textile processing, wastewater is generated. The characteristics of these wastewaters depends on the type of process but in general it produces wastewaters of great chemical complexity and diversity including many dyes and chemicals containing trace and heavy metals such as Cr, As, Cu and Zn, non-biodegradable highly persistent organics and pesticides. Therefore, due to existence of persistent organics and poor biodegradability, advanced treatment processes are required, especially when the goal is reusing the treated wastewater [92-94]. For this reason, membrane technology can be considered as an efficient candidate for providing high quality permeates.

Many investigations have been conducted to study the application of different polymeric membranes either alone [95-104] or in combination with other techniques such as electrocoagulation [105] or biological treatment in the form of membrane bioreactor (MBR) [106-111] for the treatment of textile wastewaters, and for the recovery of dyes and salts [112-116]. However, in recent decades, ceramic membrane separation has attracted significant interest to treat textile effluents because of their reliable performance even in rough operational conditions [103, 117-126]. Table 3 illustrates an overview of some studies evaluating the performance of ceramic membranes to treat textile wastewaters. From technical point of view, reasonable reduction of BOD, COD, TDS, turbidity, SS, and moderate to high rejection of dye was achieved by applying $\text{Al}_2\text{O}_3/\text{TiO}_2/\text{ZrO}_2$ ceramic membranes with MWCOs ranging from 1KDa to 500KDa. Generally, low operating pressure was observed

during experiments, while rejection rate and permeate flux could be modified by choosing the right cross flow velocity (CFV) and MWCO and operational conditions. Different methods including washing with tap, deionized, permeate, and alkaline and acid solutions were used for membrane cleaning. Around 90% flux recoveries was achieved after employing the cleaning protocols. As shown in Table 3, Alventosa-deLara et al. (2012) investigated the effectiveness of commercial multi-channel tubular $\text{ZrO}_2\text{-TiO}_2$ ceramic UF membranes in the removal of dye from the synthetic coloured feed solution. Based on their findings, at optimal operating conditions (CFV: 2.53 m/s and TMP: 4 bar) a significant dye rejection of about 95% was achieved [117]. In a similar study, Zuriaga-Agustí et al.(2014) achieved about 93% and 98.5% removal of dye and of organic matters respectively by applying tubular $\text{ZrO}_2\text{-TiO}_2$ ceramic UF membranes regardless of operational conditions [121]. Jedidi et al. (2011) employed ceramic MF membrane fabricated from mineral coal fly ash with average pore size of 4.5 μm to treat textile dyeing effluent with 3440 mg/L COD concentration. Approximately 74.5% and 99% of COD and turbidity reduction are achieved by using their proposed process [123]. Bhattacharya et al.(2010) used untreated sulphur black wastewater with 3910 mg/L COD concentration as a feed solution to study the efficiency of tubular multi-channel α -alumina and clay ceramic MF for dye and COD removal. 99% and 80% of dye and COD removal was achieved respectively [124]. Zebić Avdičević et al. (2017) achieved 98% of dye rejection by applying ZrO_2 ceramic UF with MWCO of 1 KDa [120] whereas only 62-79% of dye removal was achieved by applying $\text{TiO}_2\text{-ZrO}_2$ UF ceramic membrane with MWCOs ranging from 30 to 150 KDa [119]. The application of tubular ZrO_2 ceramic UF membrane for the pretreatment of biologically treated textile effluent prior to NF was investigated by Fersi and Dhahbi (2008) [103]. The effluent contains 329.4 mg/L of COD and 4240 mg/L of TDS (Table 3). Although the polymeric NF obtained more than 90% removal of

colour and turbidity, adding ceramic UF pretreatment resulted in steady-state operation with longer constant and stable flux compared to direct polymeric NF membrane.

Ceramic membranes have been also applied for the fractionation of the salt-dye mixtures for resource recovery from the textile effluents. Ma et al. (2017) used multi-channel tubular tight UF ceramic membrane with $\text{TiO}_2/\text{ZrO}_2$ skin layer and porous Al_2O_3 support with MWCO of 8.8 KDa and pore size of 1.16 nm to treat synthetic negative-charged dye solution with NaCl and Na_2SO_4 . Based on their results, ceramic membrane was efficient to purify high salinity dyeing effluent for salts and dyes recovery with more than 98% rejection of dye and less than 10% and 30% rejection of NaCl and Na_2SO_4 was achieved respectively [118]. Lima et al. employed ceramic membranes with average pore size of 0.14 μm and 0.6 μm to treat simulated textile wastewater sample prepared by deionized water and 0.25g/L indigo powder. About 100% percent of Indigo dye retention was observed in their experiments [122].

As an industrial example, in 2004, Societe d'Impression d'Hem(SIH), one of the major textile companies in France, installed Pall Membralox[®]ceramic ultrafiltration unit with a total filtration area of 432 m² in combination with biological treatment to treat its effluent with 10000-15000 mg/L COD concentration. The new installation was capable to recycle about 50% of the treated effluent for use as washing water for the printing machines which led to significant reduction in city water consumption consequently, remarkable reduction in operational cost [127].

As mentioned before, due to existence of persistent organic pollutants (POPs) in textile effluents which are highly persistent in terms of biodegradability, and also with large variation in produced wastewater composition in different stages, treatment of textile

effluent is a complex process and advanced techniques are necessary [93]. Technically, employing ceramic MF and UF as the pretreatment steps before NF and RO is the best option to textile wastewaters, which contains high concentrations of COD/BOD and TDS. Ceramic membrane pretreatment was found reducing the upstream membrane replacement frequency because of their superior chemical stability and resistance to harsh cleaning agents. However, high investment cost limits their application in large-scale textile plants. In perspective of dyes and salt recovery, using tight ceramic UF membranes seems to be a better option in comparison with dense polymeric NF for the rejection of divalent salts such as Na_2SO_4 . In summary, more focus need to be devoted in the development of cost-effective ceramic membranes with appropriate permeability and high filtration capacity for large scale textile operations. In fact, future research should focus on lowering the capital cost of ceramic membranes to make it more competitive to polymeric membranes. Life cycle cost assessments that includes Investment cost versus long-term operating cost of ceramic membranes is necessary to determine if installing such a large-scale ceramic membrane in potential textile plants can be a viable and profitable proposition. Coupling ceramics membranes with polymeric NF/RO membrane for the treatment of wastewaters with high organic and inorganic should be considered with design optimization to reduce the capital and operating costs.

Table 3: an overview of some investigations evaluating the application of ceramic membranes to treat effluents from textile industry

Operational Parameters	Feed Source& parameters	Membrane characteristic	MWCO/Pore size	Flux	Rejection efficiency	Cost	Ref.
TMP:4 bar Temp: 25°C CFV:2.53 m/s	Synthetic coloured solution with 50 mg/L dye concentration Conductivity (μS/cm):44.35	Multi-channel tubular ZrO ₂ -TiO ₂ Ceramic UF	MWCO of 150 kDa	255.86 L/m ² h	Significant dye rejection around 95%	-	[117]
TMP:3-13 bar	Biologically treated textile effluent from an activated sludge plant Conductivity (μS/cm) :8620 COD (mg/L) :329.4 TDS (mg/L): 4240	Tubular ZrO ₂ ceramic UF followed by flatsheet polyamide NF	UF Pore size:50 nm NF pore size:2nm	At 11 bar TMP and VRF between 1-2.77:40-45 L/m ² h	Adding ceramic UF pretreatment before polymeric NF process guaranteed steady state operation with longer constant and stable flux compared to direct NF	-	[103]
TMP:1-3 bar Temp:25°C CFV: 3 m/s	Synthetic negative-charged dye solution with both inorganic salts NaCl/Na ₂ SO ₄	Multichannel tubular tight UF ceramic membrane with TiO ₂ /ZrO ₂ skin layer and porous Al ₂ O ₃ support	MWCO:8800 Da Pore size:1.16nm	15-70 L/m ² h For TMP between 1 to 3 bar	Rejection of dye molecules: >98 % Rejection of NaCl<10% Rejection of Na ₂ SO ₄ <30% Efficient to desalinate high salinity dyeing effluent and recover salts and dyes	-	[118]
TMP:2-20 bar, Temp:30°C CFV: 3,4,5 m/s	Actual samples from a textile factory Conductivity (μS/cm) :2450-7780 COD (mg/L) :960-2525 Turbidity (NTU):35.84-83.34	Multichannel tubular TiO ₂ -ZrO ₂ UF ceramic membrane	MWCO:30,50,150 KDa	90-160 L/m ² h depending on CFV and MWCOs	Rejection efficiency (%) COD:62-79 Color:62-79 Turbidity > 99 For all MWCOs with the lowest CFV, higher removal of conductivity and COD achieved.	-	[119]
Temp:20±1 °C and 50±1°C CFV:1,2,3 m/s	Raw mercerization wastewater Conductivity (μS/cm) :75100 TOC (mg/L) :499.20 Turbidity (NTU):14.60 TDS(mg/L):20957 TSS(mg/L):100	Tubular multichannel ceramic UF 500KDa: with Al ₂ O ₃ , TiO ₂ ,ZrO ₂ active layer 2KDa: ZrO ₂ active layer, Al ₂ O ₃ support layer 1KDa:ZrO ₂ active layer	MWCO: 500, 2 and 1 kDa	Raw effluent flux with 1 KDa membrane: 29.01 L/m ² h at the beginning 28.67 L/m ² h at the end	Best rejection efficiency achieved by 1KDa MWCO at CFV:3m/s ant Temp:20°C SS:92% Turbidity:98% Color:98% TOC:53%	-	[120]
Temp:25±1 °C CFV: 3 m/s TMP:1,2,3 bar	Simulated textile wastewater sample with various CMC concentrations	Tubular multichannel TiO ₂ -ZrO ₂ ceramic UF	MWCO: 150 and 50 kDa	88.57-289.96 L/m ² h depends on MWCOs and CMC concentrations	Removal efficiency regardless of operational conditions: Organic matter:98.5% Dye:93%	-	[121]
Pressure :3 bar	Simulated textile wastewater sample prepared by deionized water and 0.25g/l indigo powder Turbidity ≥1000	Ceramic membranes	Avg.pore size diameter of 0.14μm and 0.60μm		Permeate Indigo concentration: 0 Turbidity:2-4	-	[122]
Pressure :1 bar	Textile dyeing effluent Conductivity (μS/cm) :6.16 Turbidity (NTU):45.5 COD (mg/L) :3440	ceramic MF membrane made of mineral coal fly ash	Avg.pore size diameter of 4.5 μm	100 L/m ² h	Permeate quality: Conductivity (μS/cm) :5.38 Turbidity (NTU):0.58 COD (mg/L) :880	-	[123]
TMP:0.4-1.2kg/cm ²	Untreated sulphur black wastewater Conductivity (μS/cm) :36.9 Turbidity (NTU):5912 COD (mg/L) :3910 TSS(mg/L):5550 TDS(mg/L):20200 Dye concentration(mg/L):890	Tubular multichannel α-Alumina and clay ceramic MF	Apparent porosity 36%		Dye removal :99%, COD reduction:80%	-	[124]

3.3. Petrochemical Industry

Petrochemicals are chemical products obtained from gas and petroleum processes. They generate a large volume of wastewater comprising of organic and inorganic materials with different compositions including oil compounds, dissolved formation minerals, production solids and production chemical compounds [128]. Effluents from petrochemical industry is one of the major polluter of aquatic life, and to comply with existing environmental policies and guidelines around the world, these wastewaters that contain a wide range of contaminants must be treated properly before discharging to the environment. Besides, the demand to reuse treated water has led petroleum industry to look for advanced efficient methods for treating petrochemical effluents [129].

Several research studies have been carried out to evaluate the performance of ceramic membranes in treatment of petrochemical effluents. Table 4 illustrates an overview of some studies assessing the efficiency of ceramic membranes to treat oily wastewaters. Madaeni et al. (2012) applied γ -Al₂O₃ ceramic MF with nominal pore size of 0.2 μ m to treat coke-contaminated effluent derived from a petrochemical plant with 2210 mg/L COD concentration. At 15 bar pressure and 70 °C temperature, 100% of coke removal and around 72% of COD reduction was achieved [130]. Based on the good laboratory results achieved by the abovementioned study, Salehi et al. (2014) conducted some cost analysis to evaluate economic feasibility of applying 19 channel γ -Al₂O₃ ceramic MF with 91 capacity housing unit as a pretreatment method of coke-contaminated wastewaters. The designed unit was almost 100% efficient in coke removal under operating condition of 70 °C temperature and 15 bar pressure. A total capital investment of 535300USD was estimated by the study, and break-even point (BEP) and payback period (PBP) were near 3% and 2 years respectively. This results confirmed the γ -Al₂O₃ ceramic MF unit applicability as an

488 economic potential pretreatment method for the removal of coke from petrochemical
489 wastewaters [131]. In another feasibility study, Ghidossi et al.(2009) employed an on board
490 continuous industrial scale treatment system composed of 19 channel $\text{ZrO}_2\text{--TiO}_2$ ceramic UF
491 membrane with MWCO 300kDa in treatment of oily wastewater originating from passenger
492 ships. From a technical point of view, a permeate flux of more than $100 \text{ L/m}^2 \text{ h}$ with 97%
493 hydrocarbon removal was achieved by the proposed system. From the economic point of
494 view, a satisfactory result was obtained where the cost of treated effluent by the proposed
495 system was 250000€ per year for each passenger ship. In addition, the amount of
496 wastewaters that are to be treated onshore was remarkably reduced by a factor of six. Also,
497 the investment cost which was about 70000€ per ship, covered in a short period of time
498 [132]. Abadi et al.(2011) employed tubular $\alpha\text{-Al}_2\text{O}_3$ ceramic MF with a minimum pore size of
499 $0.2 \mu\text{m}$ to treat oily wastewater samples taken from a refinery plant with 26 mg/L oil and
500 grease content. Based on the experimental results, permeate quality met the national
501 discharge standards in Iran after treatment with the proposed system where 85%, 100%,
502 and 98.6 % reduction of oil and grease content, total suspended solids (TSS) and turbidity
503 were achieved respectively [133]. Yang et al. (b) (2011), used synthetic oil in water emulsion
504 mixed with powdered activated carbon (PAC) as a feed solution to study the removal
505 efficiency and fouling behaviour of ceramic $\alpha\text{-Al}_2\text{O}_3$ MF in the presence of PAC. The study
506 showed that applying PAC had no effect on the removal of TOC with about 96% of TOC
507 removal was achieved by using oily emulsion either alone or dosed with PAC. However,
508 using PAC was an effective way to improve the permeation flux and reduce fouling as it
509 provides mechanical scouring effect [134]. In another investigation, Yang et al (a) (2011),
510 applied kaolin/ MnO_2 bi-layer composite on ceramic Al_2O_3 MF to form dynamic membranes,
511 and to enhance the separation performance in treatment of oily wastewaters. Based on the

512 experimental results, the proposed dynamic membranes achieved the best oil separation
513 performance under various operational parameters, and high permeate flux and oil
514 retention ratio of 99% was observed. However, the fabricated dynamic membranes were
515 only effective in neutral or alkaline conditions due to vulnerability of MnO_2 particles in acidic
516 conditions [135]. Mullite and mullite –alumina ceramic MF manufactured from cheap kaolin
517 clay and α -alumina powder with average pore size of $0.289\ \mu\text{m}$ is used by Abbasi et al.
518 (2010), to treat synthetic oily wastewaters with $510\ \text{mg/L}$ COD concentration. As shown in
519 Table 3, at the best operating conditions (pressure: 3bar, CFV: 1.5m/s and Temp: 35°C)
520 mullite–alumina ceramic membrane with 50% alumina content was the most suitable
521 option where 90 % removal efficiency with moderate $104\text{L/m}^2\text{h}$ permeate flux was
522 obtained. [136]. In another similar study, Nandi et al. (2010) evaluated the performance of
523 MF ceramic membranes fabricated from various cheap inorganics such as quartz, feldspar,
524 kaolin, boric acid, sodium carbonate and sodium metasilicate to treat synthetic oil-in-water
525 emulsions. By applying this self-made membrane, oil removal of 98.8% was achieved [137].
526 Despite the advantages of membrane technology over conventional treatment techniques,
527 there are some problems associated with membrane filtration of effluents generated during
528 petrochemical operations. One of them is oil droplets accumulation on the membrane
529 surface, which leads to formation of a concentration polarization layer, which consequently
530 results in permeation flux reduction. Another major challenge is membrane fouling due to
531 complex fouling characteristics of petrochemical effluents [128]. Over the past decades,
532 many investigations have been conducted to reduce membrane fouling and enhance
533 permeation flux. Surface modification [138, 139], enhancing shear stress at the membrane
534 surface by applying pulsed flow [140, 141], turbulence promoters [142, 143], vibrating
535 membranes [144, 145] and ultrasound cleaning [146] are some techniques that have been

536 used by researchers to achieve better removal efficiencies and reducing fouling and
537 concentration polarization. However, compared to the efficiency of polymeric membranes
538 either alone or as a part of MBR systems [\[147-151\]](#), ceramic membranes offer better
539 stability in petrochemical industry operational conditions. They can be cleaned more easily
540 and greater longevity. Polymeric membranes can be easily degraded and fouled during
541 treatment of petrochemical wastewaters specially when waxes and asphaltenes are present,
542 and may require regular replacement [\[12\]](#). According to Table 4, ceramic MF and UF has
543 been successfully applied for the removal of coke from coke-contaminated petrochemical
544 wastewaters. Ceramic MF and UF gives permeate of high quality with above 95% of oil
545 rejection efficiency. Moreover, ceramic MF/UF has been applied to treat one of the most
546 complex oily wastewaters called flowback and produced water generated from oil and gas
547 drilling and hydraulic fracturing operations [\[152, 153\]](#). Ceramic MF/UF is a reliable
548 technology for the treatment of produced water compared to polymeric membranes
549 because of the complex foulant profile of produced water compared to other types of
550 natural or synthetic oily effluents. Produced water contains various type of dissolved
551 minerals and salts [\[154\]](#). Veolia Water technologies employed CeraMem® ceramic
552 membranes at 60 locations to treat flowback and produced water and reuse them in the
553 shale oil operations as fracking water, reducing mains water demand [\[155, 156\]](#). However,
554 according to Ji (2015), there are only over 75 commercial ceramic units worldwide for oily
555 effluent treatment compared to more than 3000 polymeric MF/UF installations [\[128\]](#). These
556 limited number of ceramic membrane units proves that scaling up results from bench scale
557 to large scale applications is due to high capital cost of ceramic membranes specially for
558 large effluent volumes. Future studies should focus on developing precise predictive models
559 that will allow investigating the applicability of experimental results in real large industrial

operations. The main objectives of future investigations in this area should focus on improving packing densities, fabrication and coating techniques, antifouling properties, water permeability and oil separation efficiency of ceramic membranes as well as cost effectiveness [\[128\]](#).

582 Table 4: an overview of some investigations evaluating the application of ceramic membranes to treat effluents from petrochemical industry

Operational Parameters	Feed Source& parameters	Membrane characteristic	MWCO/Pore size	Flux	Rejection efficiency	Cost	Ref.
Pressure:15 bar Temp: 20-80°C CFV: 2m/s	Coke- contaminated Samples from a petrochemical company Turbidity (NTU) :251 TDS (mg/L): 265 TSS (mg/L):27, COD (mg/L):2210 BOD (mg/L) :225 VOC in 550 °C (mg/L):17	Γ - Al ₂ O ₃ based ceramic MF	Nominal pore size of 0.2 μ m	The flux was increased by increasing feed temp. 80% of flux recovery observed by proposed cleaning by NaOH	Turbidity (NTU) :13 TDS (mg/L): 32 TSS (mg/L):11 COD (mg/L):640 BOD (mg/L):55 VOC in 550 °C (mg/L):12	-	[130]
Pressure:15 Bar Temp: 80°C	Coke-contaminated effluent from a petrochemical company Turbidity (NTU) :251 TDS (mg/L): 265 TSS (mg/L):27, COD (mg/L):2210 BOD (mg/L) :225 VOC in 550 °C (mg/L):17 Coke content (wt.%): 0.1	19 channel γ -Al ₂ O ₃ based ceramic MF with 91-capacity housing	Channel diameter 4mm	Min. permeated water flux:500L/m ² h	Almost 100 percent efficient in coke removal	Economic Evaluation based on 19-channel membrane for large scale application Total capital investment :535300 USD BEP: 3% PBP:2 years	[131]
TMP :1.25 bar Temp:32.5°C and 60 °C CFV:2.25 m/s	Samples derived from a refinery plant Turbidity (mg/l) :21 TOC (mg/L): 141 TSS (mg/L):92 Oil and grease content (mg/L):26	Tubular α -Al ₂ O ₃ ceramic MF	Min. pore size of 0.2 μ m	Water flux: 500L/m ² h	Turbidity (mg/l) :0.3 TOC (mg/L): 7 TSS (mg/L): Trace Oil and grease content (mg/L):4	-	[133]
TMP:2.6-3.3 Temp:20 °C	Oily wastewater samples from passenger ships Turbidity (NTU) :150-250 Dry matter (g/L): 12-14 Conductivity (mS/cm):17-20 Hydrocarbon (ppm):35	ZrO ₂ -TiO ₂ ceramic UF membrane	MWCOs: 0.1 μ m and 300 kDa	Permeate flux: 100 L /m ² h	MWCO 0.1 μ m: Turbidity (NTU) :60 Dry matter (g/L): 9.8 Conductivity (mS/cm):17.1 HC(ppm) <1 MWCO 300 KDa: Turbidity (NTU) :59 Dry matter (g/L): 7.3 Conductivity (mS/cm):18.7 Hydrocarbon (ppm) <1	Scale up 19 channel membranes with MWCO 300-kDa composed of 19 channels was economically attractive 250,000€ per year was the cost of treated effluent by the proposed system	[132]
Pressure 3 bar Temp 35 °C CFV :1.5 m/s	Synthetic oily wastewater TSS(mg/L):60, TDS(g/L):25 COD(mg/L):510, TOC(mg/L):1000 Turbidity(NTU):89	Mullite and mullite alumina MF with 50% and 75% of alumina	Avg. pore size: 0.289 μ m	Permeate fluxes Mullite :72 L /m ² h Mullite –alumina (50%):104L/m ² h Mullite –alumina (75%):244 L/m ² h	Removal efficiencies: Mullite ceramic MF: ~ 94% Mullite –alumina (50%): ~90% Mullite –alumina (75%): ~81%	-	[136]
TMP:0.69 bar Temp:25 °C	Synthetic oil-in-water emulsion Oil concentration(mg/L):250	ceramic membrane prepared from cheap inorganic substances	82.67 percent pores with diameter between 0.1 to 0.3 μ m	19.3 L/m ² h	98.8% oil rejection efficiency	-	[137]
Pressure :1 bar Temp 35 °C CFV :4.5 m/s	Synthetic oil-in-water emulsion with powdered activated carbon P-xylene (mg/L):3000 TOC(mg/L):3817	Ceramic α -Al ₂ O ₃ -ZrO ₂ MF	Nominal pore size: 0.2 μ m	Pure water flux: 1344 L/m ² h Flux enhancement observed by dosing emulsion with powdered activated carbon because of scouring effect	96% TOC and P-xylene removal efficiency	-	[134]
TMP:1-2 bar Temp 10-40 °C CFV: 1 m/s	Synthetic oily wastewater	Ceramic Al ₂ O ₃ MF coated with Kaolin/MnO ₂	Nominal pore size: 1.0 μ m	Permeate flux 120.1 L/m ² h at 10°C 153.2 L/m ² h at 40°C	99.9% of oil retention at 10°C 98.2% of oil retention at 40 °C	-	[135]

3.4. Food Industry

The food industry consumes a massive amount of water. It is used as an ingredient of their products, in production processes, general cleaning, and sanitation and disinfection purposes. Depending on the operation processes and type of products, the volume and characteristics of these effluents can vary and sometimes difficult to predict. Generally, they are characterized by high BOD and COD plus oils, fats and nutrients. In addition, micropollutants including hormones, surfactants, antibiotics, and pesticides may be present. Consequently, effluents from food industries are a threat to the environment and appropriate treatment systems are required to remove undesirable components before discharge or reuse [157-159].

Ceramic membranes are important processes in food industry. They can be used for a variety of purposes such as treatment and clarification of effluents generated during dairy, juice, beverage, beer, wine, vinegar , sugar, olive, corn, soy sauce, meat, poultry and seafood production processes [160-176]. Ceramic MF and UF have received a lot of attention in the treatment of food industry wastewaters because of their long-term solid performance in these applications over the polymeric membranes [12]. Table 5 shows the results of some investigations evaluating the application of ceramic membranes to treat effluents from food industry. Several studies have been conducted to evaluate the efficiency of ceramic MF fabricated from natural clay and $\text{Al}_2\text{O}_3/\text{ZrO}_2/\text{TiO}_2$ materials in treatment of food industry effluents. Kumar et al.(2016) applied cheap ceramic MF membranes fabricated from clay materials with a pore size of $0.309\text{ }\mu\text{m}$ to treat raw dairy waste water with a COD concentration of 1462 mg/L . Based on their results, the COD concentration in permeate was reduced to 135 mg/L which was below the acceptable limit of permeate stream ($<200\text{ mg/L}$) [160]. As compared to similar studies done for the treatment of dairy

wastewater with polymeric membranes [177-179], good results were obtained in terms of COD removal, with 91% of COD reduction achieved by the use of this system, while for polymeric MF, UF and NF membranes the rejection rate was between 78% and 98%. However, low-pressure operation of around 2.07 bar with a steady state flux was observed during experiments using ceramic MF. On the other hand, testing with polymeric NF membranes, resulted to pressure reaching about 10 bar in some experiments. This suggests MF ceramic membranes option is more energy efficient than polymeric membrane which is a trade-off between the capital and operation costs. Hart et al. (1988) examined the possibility of employing bench-scale ceramic Al_2O_3 MF membrane for the treatment of poultry scalding and chiller waters for reuse purposes. Based on the experimental results, ceramic MF with 0.2-0.45 μm pore size was an appropriate treatment method, and the permeate quality met the standards to reuse where the permeate turbidity was less than 1NTU from the 0.2 μm filter. However, their study suggested that longer term operations using commercial scale equipment are necessary to achieve a reliable information and make a precise financial analysis [161]. Değermenci et al. (2016) applied $\alpha\text{-Al}_2\text{O}_3/\text{ZrO}_2$ ceramic MF with 0.1 μm pore size in combination with biological system as a part of a jet loop MBR to evaluate the efficiency of this combined system for the treatment of high oxygen demanding olive mill effluent. The COD and initial phenol concentrations were between 55730-91550, and 2439-4509 (mg/L) respectively (Table 5). COD and a total phenol removal of 93% and 87% were achieved respectively. The removal efficiency remained almost stable by using combined ceramic MF and jet loop bioreactor even at various hydraulic retention times [162].

Li et al.(2010) and Almandoz et al.(2010) employed $\alpha\text{-Al}_2\text{O}_3$ & ZrO_2 ceramic MF with pore sizes ranging from 0.14 μm to 0.75 μm for the treatment and clarification of raw rice wine

and corn syrup samples. Turbidity reduction between 91.2% to 99.6% and insoluble residues rejection of 63.9% to 99.8% were achieved depending upon membrane pore size which can be a reliable alternative over conventional technologies applied in the wine and corn syrup clarification [163, 164]. In a similar investigation conducted by Li et al. (2007), α -Al₂O₃ & ZrO₂ ceramic MF ceramic with 0.2, 0.5, 0.8 μ m of pore size has been used to remove bacteria from raw soy sauce sample. More than 99% of bacteria removal from raw soy sauce samples was achieved by 0.2 μ m ceramic Al₂O₃ MF membrane. On the other hand, the percentages of removing bacteria were 97.5% and 93.8% for 0.5 μ m and 0.8 μ m pore sizes respectively. The results emphasized the importance of selecting membrane with an appropriate pore size in the filtration process [165].

The efficiency of ceramic MF and UF for the treatment of effluents from the seafood processing section has also been studied. Kuca and Szaniawska (2009) used 23-channel Al₂O₃/TiO₂/ZrO₂ ceramic 150 kDa MF for the removal of protein from salted wastewater originated from fish processing operations. They reported 81% protein removal from the salted wastewater [166]. In another comparable study conducted by Afonso and Bórquez (2002), mono-tubular ZrO₂-TiO₂ ceramic 15 kDa UF has been applied for the treatment of wastewater originated from a fish meal plant [167]. The flux rate was found to be about 4 times higher than that observed in Kuca and Szaniawska experiments (Table 5); however, a total protein rejection of only 26% was achieved. It seems that the number of channels, MWCO and type of ceramic materials affects the protein rejection. In general, multi-channel ceramic MF membrane has higher packing density which is preferred over mono-channel element. Diná Afonso et al. (2004) compared the efficiency of ZrO₂-TiO₂ mono-channel tubular ceramic 15 kDa UF with multi-channel tubular Al₂O₃/TiO₂/ZrO₂ Ceramic 1KDa NF in protein removal from a fish meal plant's wastewaters. An almost similar maximum protein

rejection of 62% and 66% were observed using ceramic UF and ceramic NF respectively. However, permeate flux was approximately 1.3 times higher in the ceramic UF than ceramic NF. This led to an economic assessment for industrial scale up based on the use of ceramic UF membranes. Based on the financial analysis results (details in table 5), applying ceramic UF for the treatment of fish meal wastewaters was economically feasible for the purpose of proteins recovery [168]. Walha et al. (2011) used multi-channel TiO₂ ceramic MF with the pore size of 0.1 µm as a pretreatment step prior to polymeric NF stage to treat raw tuna cooking juices. Compared to direct polymeric NF of tuna cooking effluent, they succeeded to improve the permeation flux by nearly 3.3 times by applying ceramic MF technique. Although ceramic MF and UF membranes are not appropriate for the rejection of low MWCO matters, they can make an impact when combined with polymeric NF membranes. Such hybrid system is typical nowadays in food wastewater treatment facilities [169]. Resistance analysis and fouling behaviour for ceramic MF and UF in food industry wastewaters have been investigated by some researchers. The membrane fouling is an inevitable phenomena that occurs due to adsorption of proteins and accumulation of unwanted compounds on the membrane surface [160]. According to studies shown in Table 5, cake filtration and pore clogging are the two main mechanisms of fouling which is a normal phenomenon in MF and UF filtration processes [163-165]. Although severe fouling observed during the filtrations of some feed solutions by ceramic membranes, original flux almost completely recovered by cleaning processes utilizing sequential washing with chemical detergents and alkaline and acid solutions [161, 163, 167]. In addition, Kuca and Szaniawska (2009) realized that changes in pressure and pH in a laboratory scale have different effects on the fouling behaviour of ceramic membranes used for the treatment of salted effluents from fish processing operations. The effect of TMP ranging between 0.5 to 2

679 bar (Table 5) was negligible on membrane fouling; however, the lowest fouling observed at
680 pH equals to 9 [166].

681 Ceramic membranes have been applied practically in various sections of food industry for
682 treatment of effluents. As an example, Isoflux™ ceramic MF membranes made by TAMI
683 industries have been successfully applied to more than 50 plants around the world for
684 various purposes in dairy industry such as bacteria reduction in milk and cheese brine and
685 miscellar casein separation from milk. Isoflux™ membranes offer better selectivity
686 efficiency and higher permeation flux over polymeric membranes whereas milk fractions
687 could not successfully obtained due to large pore size distribution [180, 181]. Tetra Alcross®
688 Bactocatch and Atech Innovations GmbH products are two other manufacturers of
689 commercial ceramic membrane systems which successfully applied their products
690 worldwide for the removal of bacteria and spores from milk, polishing filtration of citrus
691 fruit juice, production of glucose from sorghum, clarifying beverages, and production of
692 sugar syrup [182, 183].

693 As mentioned before, a specific problem associated with the filtration of food processing
694 wastewater is the accumulation of unwanted compounds on the membrane surface that if
695 improperly washed may become a source of harmful bacteria [184]. The application of
696 ceramic membrane technology can guarantee food hygiene due to its ease of cleaning and
697 sterilization [185]. Moreover, ceramic membranes were successfully applied for milk and
698 other dairy productions fractionation. However, traditional polymeric membranes cannot
699 achieve similar proper results due to wide pore size distribution which causes transmission
700 of components that should be retained through the larger pores, and retention of
701 components that should be passed by smaller pores. Despite the successful application of
702 ceramic membranes in laboratory scale and some commercial systems, there is a major

703 limitation to the industrial scale implementation of ceramic membranes in food industry
704 due to their high capital cost. In addition, a better understanding of fouling mechanisms by
705 analysing the interactions between various ceramic materials and the wide variety of
706 foulants that exists in food processing effluents can result in more stable long-term
707 operations and lower operational costs [\[12\]](#).

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Table 5: an overview of some investigations evaluating the application of ceramic membranes to treat effluents from food industry

Operational Parameters	Feed Source& parameters	Membrane characteristic	MWCO/Pore size	Flux	Rejection efficiency	Cost	Ref.
Pressure:2.07-4.14 bar Temp: 25°C Cross flow rate: 5.55-22.2 m/s $\times 10^{-7}$	Raw dairy wastewater Conductivity (mS/cm): 853 Total suspended content (mg/L): 976 TSS (mg/L):254, TDS (mg/L):722 BOD (mg/L) :758, COD (mg/L):1462	Low cost tubular ceramic membrane fabricated from natural clay materials	pore size: 0.309 μm	Avg. permeate flux: 9.3 L/m ² h	COD (mg/L):135 91 % of COD removal	Membrane Cost: 69 \$/m ²	[160]
Pressure :1 Bar Temp: 15-52°C	Poultry chiller and scalding water and frankfurter chiller brine Total solids (%): 0.14 -15.7 Ash (%): 0.048-14.07 Nitrogen (%) :0.009-0.025 Fat (%): 0.02-0.05	Al ₂ O ₃ based ceramic MF	pore size: .02-.45 μm	Avg. permeate flux: 110-440 L/m ² h	Total solids (%): 0.07-14.75 Ash (%): 0.04-13.96 Nitrogen (%) :0.003-0.025 Fat (%): 0.014-0.025 Turbidity (NTU):<50 –with many less than 1	-	[161]
Pressure:0.8-2.8 bar Temp: 25°C Cross flow rate:20-65 L/min	Olive mill wastewater COD (mg/l) :55730-91550 BOD(mg/L):29930-38600 TOC (mg/L): 18620-23454 Conductivity (mS/cm): 10.04-12.01 Total phenol (mg/L):2439-4509	Jet loop MBR using α -Al ₂ O ₃ –ZrO ₂ Ceramic membranes with 37 channels	Pore size:0.1 μm	Avg. permeate flux:0.9 L/m ² h for 0.418 m ² of membrane surface area	COD removal:91-93% Total phenol removal:80-87%	-	[162]
TMP:1 bar Temp: 15 \pm 3°C CFV:1.10 m/s	Raw rice wine samples Total insoluble solids (g/L):12.7 Crude protein (g/L):10.8 Turbidity (NTU): 35.3	α -Al ₂ O ₃ & ZrO ₂ Ceramic MF	Pore size: 0.2-0.5 μm	18-33 L/m ² h	Total insoluble solids (g/L):0.7-1.2 Crude protein (g/L):2.56-4.32 Turbidity (NTU): 2.12-3.10	-	[163]
TMP:0.5 bar Temp: 60°C CFV:0.5 m/s	Corn Syrup Turbidity (g/ L BaSO ₄):1.343 Insoluble residues (%w/w):0.332 Total proteins:(%w/w):0.1	Composite tubular ceramic MF fabricated from alumina-silicate materials	Pore size: 0.5 ,0.75,0.14 μm	Permeate flux: 77.5-136.2 L/m ² h	Turbidity reduction:97.4-99.6% Insoluble residues rejection:63.85-99.75% Total Protein Rejection:70-80%	-	[164]
TMP:1 bar Temp: 22 \pm 3°C CFV:0.58 m/s	Raw soy sauce Turbidity(NTU):18.7 Total solids(kg/L):0.38 TN (g/100mL):1.74 Total bacterial count 3200	α -Al ₂ O ₃ & ZrO ₂ Ceramic MF	Pore size: 0.2,0.5,0.8 μm	3.5-12 L/m ² h Highest flux: 0.2 μm (α -Al ₂ O ₃) Lowest flux: 0.8 μm (α -Al ₂ O ₃)	Raw soy sauce permeate Turbidity(NTU):0.813-1.61 Total solids(kg/L):0.26-0.31 TN (g/100mL):1.64-1.70 Total bacterial count 30-200 >99% bacteria removed from raw soy sauce	-	[165]
TMP:0.5 to 2 bar Temp:20°C CFV:4 m/s	Salted wastewater from fish industry BOD (mg O ₂ /dm ³):2050-7980 COD(mg O ₂ /dm ³):4250-14600 Protein (wt.%):0.1-2.5	23 channel Al ₂ O ₃ /TiO ₂ /ZrO ₂ Ceramic MF	MWCO:150k Da	Flux:19.1-27 L/m ² h	Protein Rejection:81% BOD:72% COD:60%	-	[166]
Pressure: 4 bar CFV:4 m/s Temp:20 °C	Effluents from a fish meal plant	ZrO ₂ - TiO ₂ mono-tubular ceramic UF	MWCO:15 k Da	95.4 -97.7 L/m ² h	Total proteins rejection:26% Oil and grease rejection:40% Total solids rejection:4.1% UF with lower MWCO or NF is recommended for efficient protein removal.	-	[167]
Pressure :3-4 bar Temp:21 °C CFV:3-4 m/s	Effluents from a fish meal plant Total Solids (g/l) :24 Volatile solids (g/l):18.9 Total proteins (g/l) 15.5 Oil and grease (g/l) 1.21	Mono-tubular ZrO ₂ -TiO ₂ UF membranes	MWCO:15 KDa	30.1-38.9 L/m ² h	Protein Rejection:49-62%	A plant handling 10 m ³ /h of effluent: NPV:160000 USD Interest rate of return :17% Payback time: 8 years	[168]

711 Table 5: Continued

Pressure :3-4 bar Temp:25 °C CFV:3-4 m/s	Effluents from a fish meal plant Total Solids (g/l) :24 Volatile solids (g/l):18.9 Total proteins (g/l) 15.5 Oil and grease (g/l) 1.21	19 channel tubular Al ₂ O ₃ /TiO ₂ /ZrO ₂ Ceramic NF	MWCO:1 KDa	22.3-32 L/m ² h	Protein Rejection :63-66%	[168]
MF: Pressure:2 bar Temp:25 °C CFV:3.1 m/s NF: Pressure 35 bar Temp:40°C CFV:2.5 m/s	Raw tuna cooking juices Turbidity(NTU):496±7 SS(g/L):2.1±0.1 COD (g/L):23.5±0.7 Dry matter(g/L):147.7±3	Multi-channel TiO ₂ ceramic MF followed by polymeric NF	MF Pore size :0.1µm	MF/NF :90-100 L/m ² h NF:30 L/m ² h	Retention of inorganic compounds:70-76 % Permetation flux significantly increased by applying ceramic MF pretreatment	[169]
TMP:1.75 Temp:50 °C	Rinsing water from bottle washing machine COD (mg/L):240-580	Al ₂ O ₃ Ceramic MF	Pore size :0.2µm	40 -160 L/m ² h	Turbidity retention :99% COD rejection:30%	[170]

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3.5. Pharmaceutical

Water is an inevitable component in pharmaceutical manufacturing operations, and it is widely used in different stages of processing, from formulation to production of drugs for use as medications. The wastewaters generated in different stages contain a wide variety of organic and inorganic compounds and impurities. In addition, removal of pharmaceutically active compounds (PhACs) and endocrine disrupting compounds (EDCs) from generated wastewater is essential and has become one of the major concerns in recent years. Improper and insufficient treatment of pharmaceutical effluents may cause PhACs return to human body through water cycle which can lead to irreversible consequences. EDCs are also known to disrupt the human endocrine system [186, 187]. It is estimated that many wastewater treatment systems are not designed to remove specific compounds, making them inefficient at removing pharmaceutical compounds before effluents are discharged to the environment. Therefore, the Pharmaceutical industry requires a robust and high quality wastewater treatment system to meet the discharge limits. [188].

The application of MF, UF and NF polymeric membranes either alone or as a part of MBR has been studied in pharmaceutical industry for various purposes. These include removing of organic compounds and endocrine disruptors from the pharmaceutical effluent, separating and recovering of antibiotics from the pharmaceutical wastewater, and isolation and purification of biologically active compounds such as viruses and enzymes [9, 189-199]. However, polymeric membranes are sensitive to aggressive cleaning agents, so they cannot be efficiently sterilized and should be replaced at more frequently.

The applicability of ceramic membranes is increasing in pharmaceutical industry due to its better capability in terms of repeated steam sterilization and ease of cleaning with harsh chemicals compared to polymeric membranes [8, 200]. Hydro Air Research Italia membrane

systems are operating successfully for antibiotic recovery by using ceramic membrane in fermentation broth clarification step before multistage RO which ensures excellent protein removal and high permeate flux [201]. GlaxoSmithKline a British pharmaceutical company replaced polymeric membranes with Star-Step™ ceramic membrane system supplied by Mantec filtration. It consists of 4 banks of 8 housing with 108 ceramic membrane unit in each housing of 208 m² membrane area at its antibiotic drug processing plant. Several economic advantages such as less maintenance requirement, longer membrane life span, energy saving by using star-shaped flow channels and easier membrane cleaning were observed by replacing polymeric membranes with ceramic units that led to the reduction in operating costs. From the technical point of view, the ceramic unit represented remarkable filtration performance, doubling the flux of previous polymeric membrane system [202]. Inopor GmbH, one of the suppliers of ceramic membranes for the purpose of liquid filtration, applied ceramic UF in the treatment process for “water for injection” in pharmaceutical industry. Ceramic UF has to be installed to prevent the growth of microorganisms that would lead to contamination of piping system and the purified water after electrode ionisation stage in pharmaceutical industries [37]. Polymeric membranes are not appropriate for this purpose due to low resistant against organism and disinfection by steam. Inopor® ceramic membrane showed excellent efficiency for this requirement [37].

In summary, application of ceramic membranes in pharmaceutical section have the benefits of being frequently bio inert, persistent against bacteria and can withstand repeated chemical and steam sterilization at high temperatures which polymer membranes may fail to tolerate [6, 200]. Employing ceramic UF as a part of hybrid system for generating ultrapure “water for injection” for medical liquids including heat exchange, reverse osmosis,

membrane degasification and electrode ionisation can appropriately prevent the growth of microorganisms and pollution of piping system where the application of polymeric membranes is not possible due to poor resistance against periodic steam sterilization. However, very selective concentrations and recovery of vitamins, enzymes and antibiotics is another subject of interest in pharmaceutical industry and most of the ceramic membranes show poor selectivity properties. Future research in this industry sector may focus on introducing new polymer-ceramic composite membranes which can guarantee high selectivity and great tolerance to aggressive conditions. Introducing new polymers with high selectivity for the purpose of coating on ceramic nanofilters with lower MWCOs below 450Da maybe an efficient way for complete separation of pharmaceuticals with complex structures out of wastewater [8, 37].

3.6. Mining Industry

Acid mine drainage (AMD) is the most common water pollution issue in the mining industry. It is produced when rock containing sulfur-bearing minerals is exposed to oxygen and water. Typically, AMD is characterized by low pH, high specific conductivity and high concentrations of heavy metals and other toxic elements [203-205]. If AMD is left untreated and gets to nearby water systems such as rivers, streams or lakes, it can contaminate surface and groundwater and may disturb the reproduction system of aquatic life. It also affects metal and concrete structures by corrosion, and can raise water treatment costs. Consequently, the development of cost-effective and efficient treatment solutions for the AMD problem has been the subject of many researches during recent years.

Some investigations have been published on wastewater treatment and recovery of heavy metals from the mining effluents by various pressure-driven polymeric UF/NF and RO

803 systems [\[206-219\]](#) , and has shown that polymeric NF and RO have quite similar efficiency in
804 the removal of heavy metals from mining effluent. However, applying polymeric NF due to
805 higher flux, acceptable rejection efficiency and, lower energy consumption was more
806 appropriate than RO at comparatively low-temperatures. On the other hand, using RO is
807 preferable over polymeric NF at high temperature conditions [\[208, 217, 219\]](#). Membrane
808 fouling, however, is the major challenge for using polymeric membrane in harsh
809 environment such as mining wastewater treatment. In contrast, ceramic membranes may
810 be an appropriate alternative over polymeric membranes due to their significant chemical,
811 mechanical and thermal stability under harsh operating condition like what is expected in
812 actual mining operations. As an example, Blackhawk Colorado AMD treatment facility
813 replaced tubular polymeric membranes with ceramic membranes in 1995, and were still in
814 service till 2013 which proved the durability of ceramic membranes compared to polymeric
815 membranes lifespan ranging between 6 to 9 months [\[220\]](#). Another successful example of
816 commercial application of ceramic MF system is the new wastewater treatment plant
817 installed in 2009 at the Upper Blackfoot Mining Complex located in Montana, USA.
818 Compared to high density sludge clarifier system installed at this facility, ceramic MF could
819 operate at very acidic condition which is an advantage for AMD treatment. Lower pressure
820 operations, smaller footprints, lower chemical consumption, labour and power costs were
821 some significant improvements achieved by replacing the clarifier system by ceramic
822 MF[\[220, 221\]](#). Liqtech International Inc. installed fifteen particle removal and six dewatering
823 systems using CoMem® SiC ceramic membrane with 25 and 146 mm outer diameter (OD) in
824 one of the largest mining operations in Europe. Particle removal system was placed in a 99X
825 glassfiber reinforced plastics multihousing in racks of 6 housings, and the dewatering system
826 was placed in a single polypropylene housing in racks of 6 housing. The actual membrane

827 capacity was 960 m³/h and the proposed system could remove heavy metals as required
828 and stable permeability and steady state permeation flux were observed during operations.
829 However, there is no information available in supplier website regarding cost analysis [222].
830 In addition, based on Mine Waste Technology program 2002 annual report, Stewart
831 compared investment and operating cost of 1136 L/min polymeric and ceramic membrane
832 systems. The capital cost of 1136 L/min ceramic membrane system was about 1900000 USD
833 while for the polymeric membrane system with the same capacity, the capital cost was
834 about 1800000 USD which was only about 5% cheaper compared to ceramic membrane
835 system. On the other hand , the annual operating cost of 1136 L/ min ceramic membrane
836 system was about 55 % less than the similar capacity polymeric membrane system due to
837 lower maintenance cost of ceramic membranes which is a big difference (about 260000 and
838 470000 USD/year for ceramic and polymeric units respectively) [220].

839 A review of previous commercial scale application of ceramic membranes in treatment of
840 harsh mining effluents proves that ceramic membranes afford specific advantages in this
841 application like long-term durability, ease of cleaning, better chemical and mechanical
842 stability and lower annual operational cost compared to polymeric membranes. However,
843 the main disadvantages of ceramic membranes are high fabrication costs of ceramic
844 components and high capital cost associated with installation of commercial scale ceramic
845 membrane units in mine sites [6]. Future development should be based on new ceramic
846 components with reasonable fabrication cost and appropriate permeability which can
847 handle stable and steady state operations even under harsh conditions faced in mining
848 operations and more space-effective ceramic membrane modules with smaller footprints [8,
849 12]. The main problems associated with the treatment of AMD are managing the large
850 amount of sludge, which arises during treatment process, and various chemical processes

with complicated behaviour and various reaction rates in AMD. A design an appropriate hybrid system by combining ceramic MF/UF and other chemical and mechanical techniques such as electrocoagulation and high shear reactors for reducing treatment sludge rates of chemical reactions in AMD should be considered in future researches [220, 223, 224].

4. Ceramic Membrane Reactors for Advanced Oxidation Processes (AOPs)

AOPs are a group of chemical oxidation processes designed to remove natural organic compounds (NOM) and decompose of persistent non biodegradable organics in wastewater through reactions with hydroxyl radicals (OH). OH generation is possible through different techniques including ozone, H₂O₂, ultraviolet (UV), Fenton reaction, electrolysis and photocatalysis [225].

Although membrane filtration is an effective technique for removal of wide range of toxic elements from the wastewaters, it has limited ability to remove some dissolved organics, especially in the range of MF/UF. Therefore, combined membrane separation with AOP in a reactor as a single system looks like an appropriate solution to provide flexibility and improve the limitations of both AOPs and membranes [225, 226]. However, one of the major drawbacks associated with advanced oxidative membrane reactors is conflict of hydroxyl radicals and membrane polymers [225]. Consequently, researchers have tried to develop various ceramic membrane reactors for AOPs to overcome the Incompatibility issue between hydroxyl radicals and membrane polymers. In addition, ceramic membranes are more resistive to UV radiation whereas polymers may deteriorate easily [226-228].

Ozonation, which refers to the application of ozone in wastewater treatment, is one of the advanced oxidation processes involving the production of very reactive oxygen species that

are able to attack and degrade a wide range of organic pollutants in the wastewaters [226, 229]. This method has been successfully combined with ceramic membrane filtration process for more effective treatment of industrial complex wastewaters and efficient removal of organic toxics. Majority of investigations has been applied ozonation either as pre-treatment stage for removal of toxic organics prior to the membrane filtration step [230, 231] or post-treatment stage to treat both permeate and retentate [232-237]. However, studies about advanced ozone ceramic membrane reactor as an integrated process is limited in literature. A few works successfully demonstrated the use of inorganic membranes and ozonation in a hybrid reactor to achieve higher TOC removal with minimum amount of ozone usage [238-240]. Further work is required to address the high energy consumption and toxicity issue associated with ozone generation [229] to make this method an economic and reliable alternative for removal of organic pollutants from industrial effluents in large-scale operations. Figure 4 illustrates an ozone membrane reactor. Numbers 1 and 2 on the Figure Indicate alumina capillary membrane and zeolite membrane unit respectively.

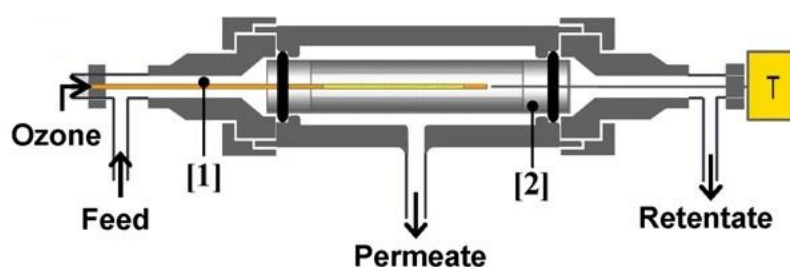


Fig 4. Ozone membrane reactor (reproduced from [241] with permission from the Journal of Membrane Science)

896 Researchers widely studied reactor configuration that is called photocatalytic ceramic
897 membrane reactors (PMRs). It consists of a ceramic membrane module as a second stage
898 and photocatalytic reactor as an OH generator in an integrated hybrid process. Successful
899 laboratory-scale applications of this method in the removal of organics including
900 pharmaceutical pollutants and drugs from wastewaters have been reported by [\[242-250\]](#).
901 Studies were mostly done in a laboratory scale because of high cost of UV source. Finding
902 and developing cost effective alternatives to replace UV such as sun as a cheap source of
903 light to generate efficient OH radicals will allow the potential of scale up for industrial large
904 scale operations [\[251\]](#). Additionally, mechanisms of transformation of organics in PMRs and
905 the possibility to separate the reaction zone from the separation zone is not fully realized
906 so far. Comprehensive studies are still required to determine these kind of mechanisms
907 which have a great significance on the overall system performance[\[226, 251\]](#). Innovation in
908 the usage of nanoparticles immobilized in ceramic materials can further prevent
909 photocatalyst degradation cause by irradiation and leads to a continuous operation in the
910 system[\[251\]](#). Figure 5 shows a schematic diagram of photocatalytic membrane reactor
911 system with immersed UV-A lamps which is used in Sarasidis et al.(2014) experiments [\[249\]](#).

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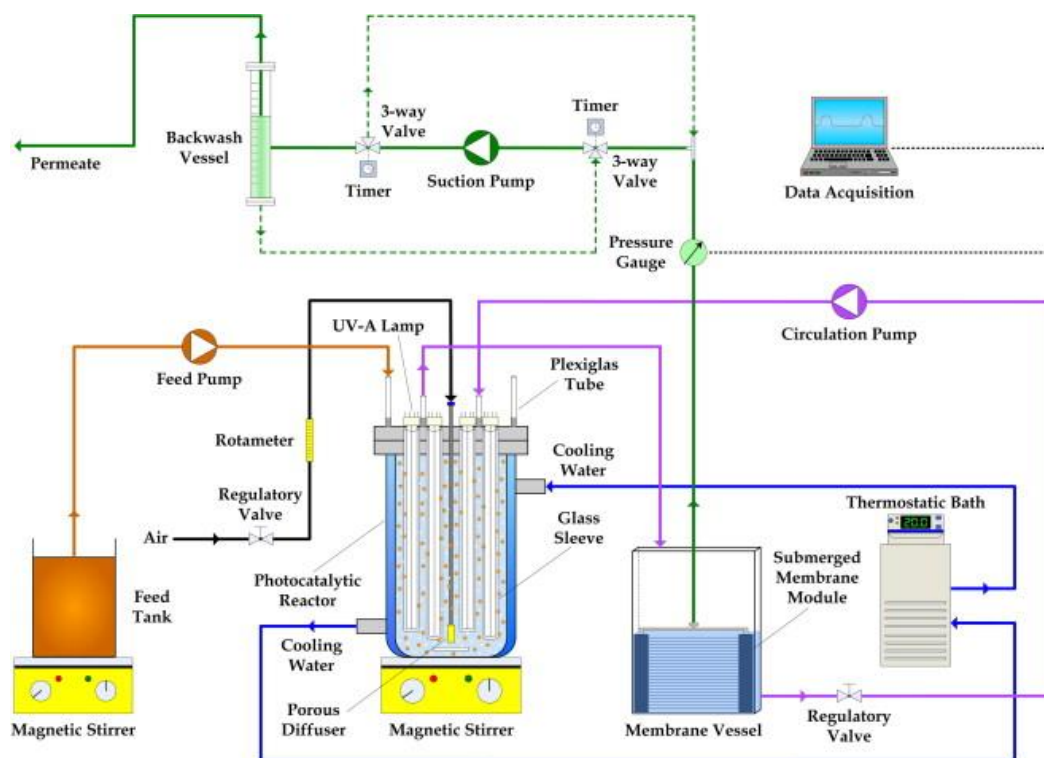


Fig 5. Photocatalytic membrane reactor with immersed UV-A lamps (reproduced from [249]

with permission from the Chemical Engineering Journal)

Another promising combined configuration is electrochemical advanced oxidation processes (EAOPs) integrated with membrane modules as a hybrid reactor. Actually, the membrane unit has two roles in the system. It acts as electrode for electrochemical process and membrane for filtration [226]. From technical and economic point of view, this process seems to be a better choice compared to PMRs. it is capable of complete mineralization of persistent organics and production of high quality effluent because of generation of large quantities of hydroxyl radicals during the electrolysis. In addition, reactive electrochemical membranes do not require UV for OH generation and relatively inexpensive unlike PMRs [226]. However, industrial adoption of this technique depends on improved understanding of factors such as influence mechanism of organic and inorganic ions on performance of electrochemical membranes. Optimisation of operating parameters such as permeate flux, pressure, flow rate, pH and temperature leads to maximizing the efficiency of the hybrid

system[[226](#)]. Introducing cost effective doping techniques to produce ceramics with high electrical conductivities seems to be a promising way for the production of electrode materials with high mechanical resistance which efficiently generate high yields of hydroxyl radicals [[252-254](#)].

5. Ceramic Membrane Cost analysis

It is very important to consider the type of application and fouling potential of feed wastewater for economic analysis of ceramic membranes. In terms of capital expenditure, commercially available ceramic membranes are three to 10 times more expensive than the polymeric membrane modules and hence many companies prefer polymeric option in their new installations [[255](#), [256](#)]; however, in aggressive operational environment the ceramic membrane may be a better alternative for wastewater filtration because of the remarkable reduction in operational costs. Normally the operating cost of an industrial scale membrane system is originated from the power and energy cost required to maintain a steady state long term permeate flux, the cost of membrane replacement, the cost of chemical and reagent required for membrane cleaning, and some of the costs such as worker's wages. Longer life span, ease of cleaning by high temperature steam sterilization and other potential unconventional method such as using electric and magnetic fields, and the ability to recover initial permeability and water flux by back flushing and proper cleaning, are all reasons that ceramic membranes have reduced operating cost than polymeric membranes [[12](#)]. Nanostone Water Inc. developed a ceramic versus polymeric cost model to estimate a total of 10 years' operating expenses based on pilot data collected from various industrial wastewater reuse plants using UF/MF membranes. Overall, the ceramic membrane reduced operating costs by 55 % [[257](#)]. Despite superior characteristics of ceramic membranes

including high chemical, thermal and mechanical stability, ease of cleaning with various methods and longer lifespan, the high material cost of ceramic membranes which is around 500-2000 USD/m² compared to 50-400 USD/m² of polymeric membranes [255, 258-262], is the main drawback of applying this system in large scale industrial applications. The focus of future research should be in the development of cheap ceramics membrane for industrial applications including cheaper manufacturing and material costs. This could be achieved by incorporating new nanomaterials in membrane manufacturing technology. Utilizing engineered nanoceramic materials with improved hardness and mechanical and thermal strength for use in ceramic matrix composites will enhance the performance of ceramic membranes technically and economically. However, industrial scale application of nano-enabled ceramic membrane technology by industry may take years [263] because a lot of research need to be done to improve the current sintering methods [38].

6. Summary:

Ceramic membranes have outstanding features over polymeric membranes because of their remarkable robustness, ease of cleaning and high membrane life. However, there are still challenges in the application this technology for industrial effluents filtration that needs to be addressed in future researches. From the previous literature review conducted, it is deduced that the number of industrial scale application for the treatment of industrial effluents by ceramic membranes in comparison with laboratory investigations is very limited. This is due to its high capital cost, which encourages the industry shareholders to use polymeric systems for their liquid filtration purposes. Reducing ceramic membranes investment cost by introducing new fabrication processes will most likely make ceramic membranes an economically competitive alternative to polymeric membranes for many

industrial scale applications. In the future, developing Ceramic nanofilters with lower MWCOs and with the ability to control pore size distribution may offer significant advantages in certain operating cases such as recovery of valuable materials in various industrial sections. Introduction of advanced sintering methods with low sintering temperature and short sintering times for the fabrication of nanoceramics can prevent the decomposition of ceramics, which will consequently lead to enhancing mechanical properties of ceramics as well as financial saving. Extensive studies are still required to improve AOPs by ceramic membrane reactors. Minimizing toxicity issue associated with ozone generation, developing cost-effective ceramic membranes with high resistance against irradiation to apply in photocatalytic ceramic membrane reactors, and producing ceramics with high electrical conductivity to apply as reactive electrochemical ceramic membranes in a hybrid reactor are some suggestions for future research and development in this category.

From the review of past studies, it can be concluded that the number of investigations done by ceramic membranes in the mining and pharmaceutical sections is more limited in comparison to the other industry sectors. This may be due to the complex nature of pharmaceutical wastewaters and AMD. Future research efforts should focus on further investigation of the application of various ceramic membranes with different pore sizes either alone or in combination with other physico-chemical techniques in treatment of pharmaceutical and mining effluents to guarantee steady state long-term operations without any or minimum requirement for membrane replacement.

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