

1 **Ultrafiltration membranes for wastewater and water process engi-**
2 **neering: a comprehensive statistical review over the past decade**

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12
13 **Abstract**

14 The primary intention of this review is to showcase and quantify the level of research interest and
15 current research trends, concerning UF membrane applications and processes within the past dec-
16 ade (2009–2018). Detected statistics manifested a resurgent interest in the UF technology on a
17 yearly basis. "Journal of Membrane Science" and "Desalination and Water Treatment" were the
18 primary journals dominating the size of the annual publication among more than 120 ones, with
19 854 and 683 papers, respectively. Based on ScienceDirect research platform, fouling (27%), mod-
20 elling (17%) and wastewater (12%), were the dominating research topics and counting for more
21 than half of total scientific articles published (4547 articles) within the specified period of the
22 research. Unsurprisingly, topics like UF membrane fabrication and modification, food processing,
23 hybrid membrane process have disclosed a distinguished growing up trends in terms of annual
24 publications. The current review unveiled the present-day significance of the UF membranes
25 along with their prospective opportunities for attaining sustainable water industries and material-
26 izing the efforts of future researchers into the right orientation.

28 **Highlights:**

- 29 • Fouling, modelling and wastewater are dominating research areas of UF membrane which counted for 27%,
30 17%, and 12% of the total publication's size, respectively.
- 31 • Journal of Membrane Science was the primary journal dominating the size of the annual publication about
32 UF.
- 33 • Fouling of the UF membrane is the largest single area of research interest.
- 34 • Potential research trends in UF membrane applications are critically reviewed
- 35 • Topics of Optimization and Hybrid UF membrane processes are getting a consistent research interest.

36

37 **Keywords:** Ultrafiltration, UF membrane fabrication, Research trends, statistics, UF membrane fouling.

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55 **1 Introduction**

56 The sustainable exploitation of water resources is the backbone for the sustainable evolution of
57 modern society and economy. Along with the rapid development of economy and society, more
58 pressure on the water resources deficiency due to industrialization and human activities has been
59 generated. Diversified techniques have been emerged to clean and renovate polluted waters for
60 industrial, agricultural and human being consumptions. Since their first industrial outset in 1970
61 for electrophoretic painting, Ultrafiltration (UF) membrane has come a long way as a safe, clean,
62 economical and potent separation tool for a wide range of constituents and contaminants in water
63 and wastewater.

64 Serving as an outstanding separation technique for more than one century, Ultrafiltration (UF)
65 has been harnessed in membrane filtration to mechanically separate materials from a mixture. The
66 term "Ultrafiltration" was first introduced by Benchold in 1907, forcing solutions at several atmos-
67 pheres through a membrane [1]. More precisely, the hydrostatic pressure forces induce movement
68 of a liquid to pass through a semipermeable membrane. This separation process targets molecules
69 that contain a higher molecular weight and suspended solids depending on the molecular weight
70 cut-off (MWCO) specified by the specific membrane along with other factors that can take a sub-
71 stantial role, such as molecule shape, charge and hydrodynamic conditions [2]. The main mecha-
72 nism used for UF is size exclusion, however, depending on the compounds present; reactions be-
73 tween the particles and the membrane might prevent the maximum efficiency of the process.

74 Market dynamics have led to a surging interest in UF applications, as has been witnessed during
75 the past few decades. Looking for an efficient selective separation technology with low capital
76 cost and longer membrane unit lifespan are some of these dynamics. Steadily, this UF market size
77 trend has risen with the growing environmental awareness concerning water/wastewater treatment
78 technologies, decreasing freshwater resources along with the sustainability policies and stringent
79 regulatory. The UF market was estimated at USD 950.0 million in 2017 and is projected to hit
80 USD 2,140.1 million by 2023. The Dow Chemicals, Koch Membrane System, PennWell Corpo-
81 ration, GE Corporation, Oasys Water is the top 5 global UF membrane companies in the industry.
82 The Koch and Dow Chemical Membrane System dominating the global market with over 51%-
83 unit volume share. Approximately 65% share of the global membrane market was dominated by
84 the U.S. and Asia Pacific region together mainly due to the rising demand in pharmaceutical,
85 chemical processing, wastewater management etc [3].

86 UF is an advanced separation technology employed across various industry verticals. It was
87 initially established as a fractionation technique in the late 1960s. Since then, UF membranes have
88 been continuously improved, and its applications have crossed a wide variety of fields, from chem-
89 ical recovery, cell harvesting, dairy production, medical use, wastewater reclamation, water treat-
90 ment and juice concentration [4–8]. Particularly, it is well known as a clarification and disinfection
91 separation process has a wide range of applications [9]. The focused applications for this mem-

92 brane technique lie on the purification and concentration of macromolecules, such as protein so-
93 lutions in the food industry [10–12]. Other common applications that require the use of ultrafiltra-
94 tion techniques lie in the wastewater sector, fouling, bacteria and virus removal, paint treatment
95 for the metal industry and the textile industry [13–17]. The reasons why UF replaces conventional
96 purification and disinfection process are based on the simplicity and overall cheap process due to
97 low energy usage, fewer control methods, no or less emphasis on chemicals of the process, mild
98 operating temperature and high-quality treatment. At last, introducing technology enables many
99 industries to become eco-friendlier by facilitating the recycling of waste materials and resources
100 recovery [18,19].

101 With so many journals, papers, articles and documents being presented every day on specific
102 engineering or science topics, it is a very decisive matter for researchers to have a general under-
103 standing of what has already been published so that their work does not overlap, or they find them-
104 selves doing unnecessary experiments that have already been carried out. That is where literature
105 reviews play a role; they are scholarly papers that involve current knowledge including substantive
106 findings, as well as theoretical and methodological contributions to a particular topic [20]. They
107 are not to report new or original findings but are secondary sources, usually preceding any sections
108 of work. Therefore, this statistical review, about UF technology, has been undertaken for high-
109 lighting and tracking various research trends concerning what has been accomplished in water,
110 wastewater and many other applications over the past decade. Along with anticipating research
111 trends and where should be our research prospects in the coming decade.

112

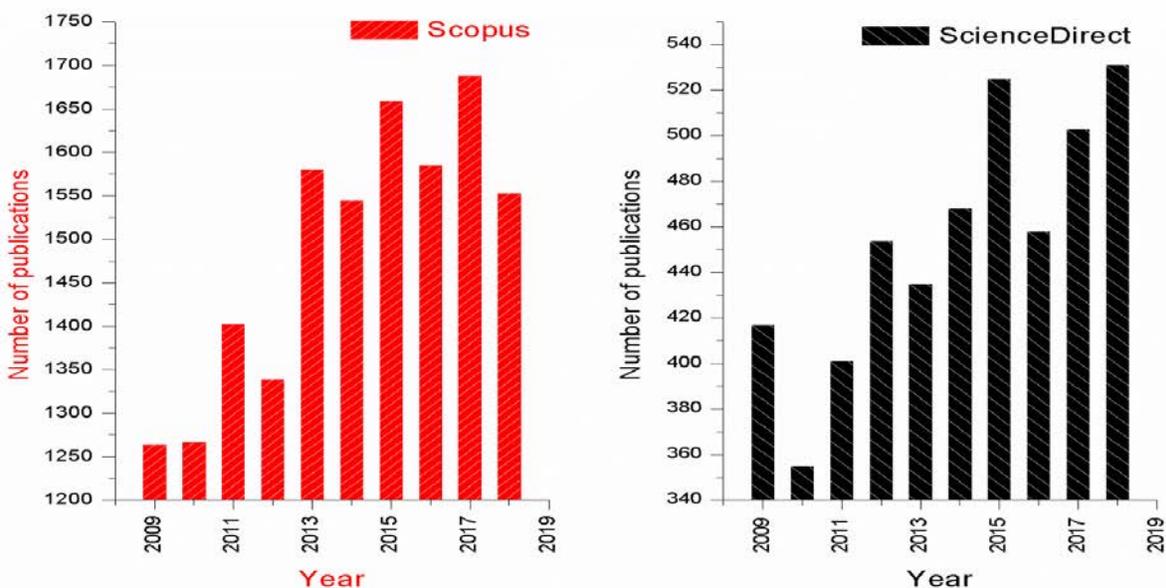
113 **2 Publications on Ultrafiltration over the past decade**

114 With the rapid developments in polymer chemistry, doping, fabrication techniques, modelling as
115 well as applications of membranes as an alternative technology, UF has affirmed their position as
116 an eminent area of research. Virtually, the exact number of articles published was tricky to deter-
117 mine, especially with differing results obtained depending on the database interrogated. In the past
118 few recent years, several statistical review articles have emerged in literature for unlike topics and
119 disciplines [21,22]. Tober (2011) has compared four popular search engines PubMed/MEDLINE,
120 Scopus, ScienceDirect and Google Scholar to assess which search engine is most functional for
121 literature research in laser medicine according to the criteria, recall, precision, and importance.
122 Results disclosed that the most efficacious search engine for an overview of a topic is Scopus,
123 followed by ScienceDirect. A more detailed study can be found elsewhere [23].

124 Herein, the ScienceDirect database was harnessed as a scientific platform for detecting the re-
125 search statistics across the spectrum of available journals. The search was specified to only involve
126 academic articles and reviews that virtually related to term "Ultrafiltration". The later processing
127 of data has eliminated any article or review that encloses text e.g. 'ultrafiltration' but does not
128 virtually fit to the respective field. For instance; the term ultrafiltration may appear rightfully in
129 the manuscript while specifically, the main theme was identified dealing with nanofiltration. At

130 this step; such articles have been discarded from the data aiming to only encircle relevant articles
131 to the UF. The general research trend of the UF membrane persisted to manifest a surge in the
132 number of academic articles published annually over the period studied for both platforms (Scopus
133 and ScienceDirect) (Figure 1). This was due to the versatility of UF membranes have been reported
134 in literature covering plentiful topics, as some will be discussed in the later sections.

135 Scopus is the world's largest abstract and citation database of peer-reviewed research literature.
136 It contains over 20,500 titles from more than 5,000 international publishers. Scopus database de-
137 livers the most comprehensive view of the world's research output in the versatile fields of tech-
138 nology, science, social science, medicine, and arts and humanities. When Scopus was interrogated
139 using "Ultrafiltration" as a search term in the title, abstract or keywords, it identified 14,882 articles
140 in the period of interest. As shown in Figure 1(Left), there is a general trend of increasing research
141 output, with 1264 publications in 2009 rising to 1402 in 2011, 1580 in 2013, 1659 in 2015, 1688
142 in 2017 and 1553 in 2018.



143 Figure 1: Yearly publications on Ultrafiltration identified by Scopus (Left) and ScienceDirect (Right).

144
145 In the meantime, Elsevier ScienceDirect delivers over 12 million publications from over 3,500
146 journals. When the search was repeated relying on ScienceDirect database, it identified a total of
147 4,547 publications, about one-third the number given by Scopus. The number of ultrafiltration
148 publications by year in ScienceDirect, as shown in Figure 1 (Right). The same general increase in
149 research output can be seen, from 417 papers in 2009 increasing to 454 in 2012, 525 in 2015, and

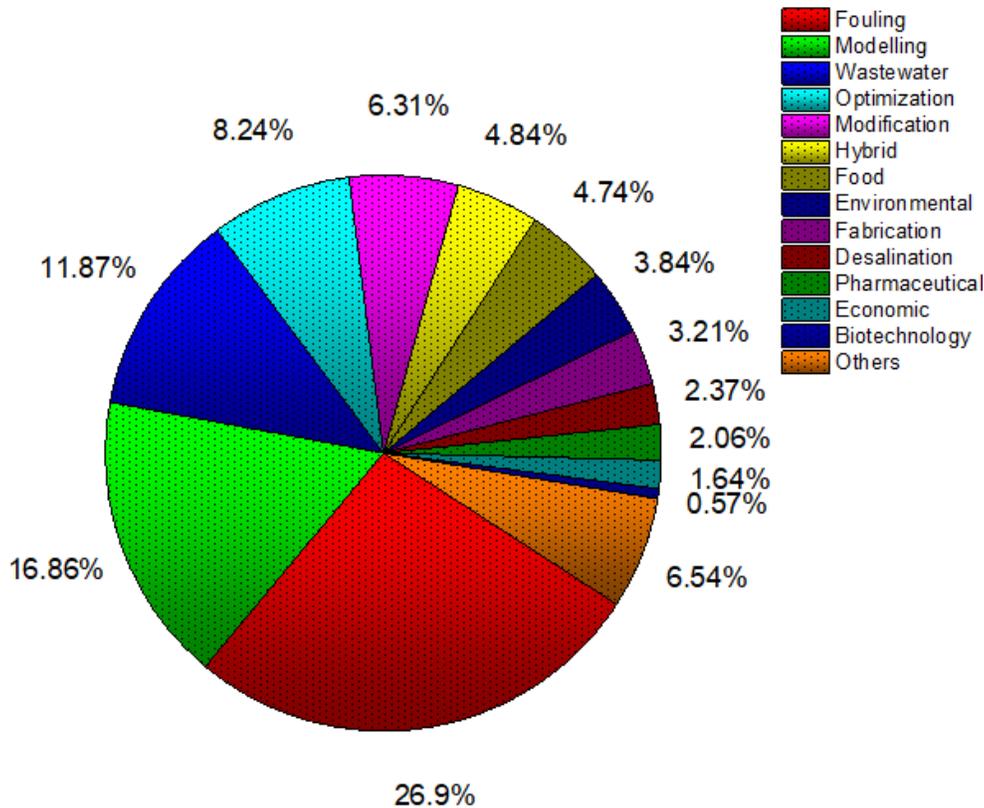
150 531 in 2018. Herein it should be noted that even though both Scopus and ScienceDirect are scientific literature databases owned by Elsevier, but ScienceDirect hosts the Elsevier content as full text whereas Scopus comprises only abstracts and citation statistics concerning both Elsevier and non-Elsevier content. And that is what could explain the variation in the number of articles been published by both databases.

155

156 3 General research trends since 2009

157 As depicted in Figure 1, a general increasing trend in UF research can be seen since 2009. Approximately 4763 publications have been released within the specified research time. These are dealt with review articles, research articles, book chapters, conference abstracts and reviews. Among these, research articles were covering more than 95% of the total publication types.

161 To elucidate the general area of research on UF membranes and processes, areas of research specified for this study were used as additional search terms in the title, abstract or keywords when searching ScienceDirect.

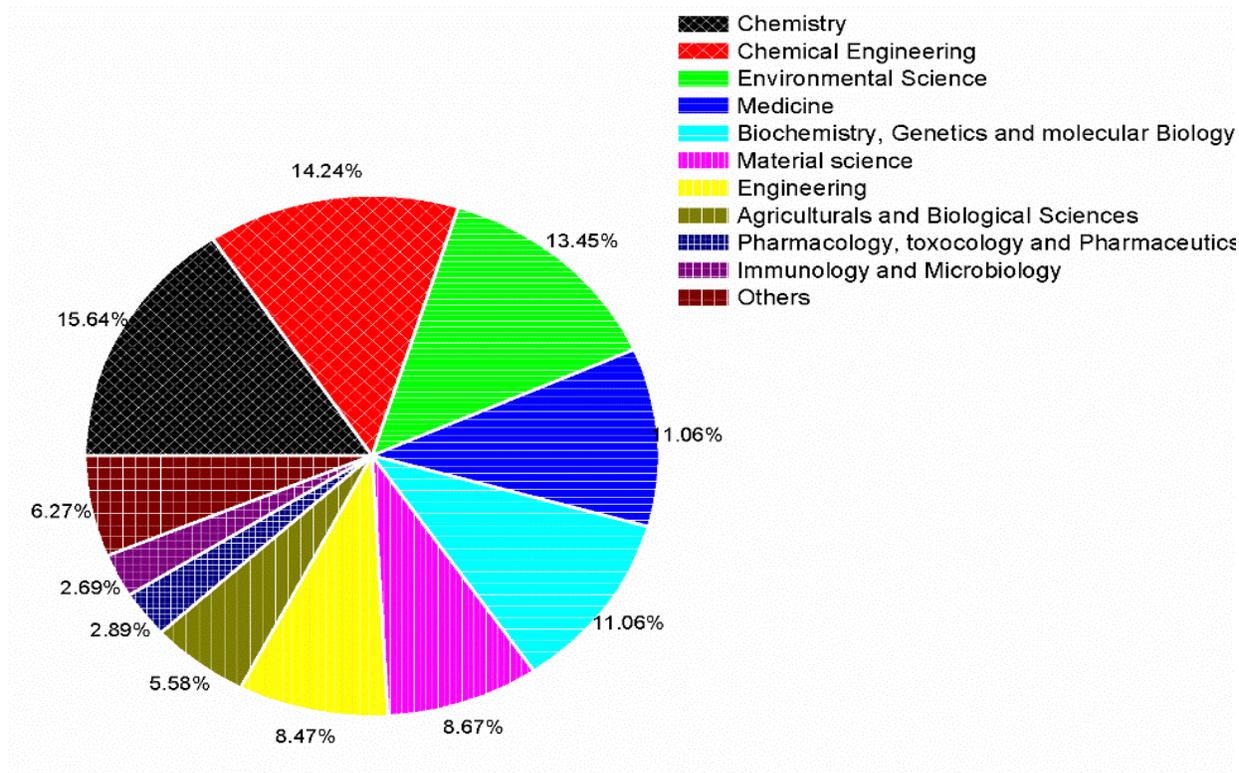


164 Figure 2: Articles identified by additional keywords in ScienceDirect (2009-2018).

165

166 Topics breakdown of articles from the total number of publications during the period between
167 2009-2018 is highlighted in Figure 2. The majority of the articles reviewed have dealt with fouling
168 applications, with 1407 papers identified, this may, however, include publications that also cover
169 other topics such as wastewater, membrane fabrication or modelling publications that have been
170 devoted to mimic fouling behaviour of membranes. Membrane fouling applications total some
171 27% of the papers reviewed, the trend is then: modelling (17%), wastewater (12%), process opti-
172 mization (8%), membrane modification (6%), hybrid membrane processes (5%), food (5%), envi-
173 ronmental studies (4%), membrane fabrication (3%), desalination (2%), pharmaceutical (2%),
174 while the rest of the search terms returned about 9% for other applications. Given that wastewater,
175 environmental and desalination can befall in one section, given by "water processing", and since
176 membrane modification is a subset of membrane fabrication and can be grouped together [21],
177 water processing and membrane fabrication/modification would be the second and third research
178 topics with 18% and 9% of total reviewed papers, respectively.

179 UF has a broad scope of applications, with research conducted out in the last ten years touching
180 on a diverse range of subject areas. The top ten subject areas account for almost 93.7% of the total
181 number of articles, with chemistry being the most active subject area for research, accounting for
182 15.6% of the total, followed closely by the subject of chemical engineering. The remaining 6.3 %
183 of papers are from subject areas such as energy, physics and astronomy, ...etc. Figure 3 elucidates
184 articles published for each of the top ten subject areas, as a percentage. It can be observed that the
185 subject areas endue significant potential crossover–developments in polymers harnessed in mem-
186 brane construction would be published in the field of chemistry and chemical engineering as well
187 as environmental science, as these three subjects are covering 43.5% of the total articles. Many
188 other subject areas indicate research into applications of UF, with medicine, biochemistry and
189 material science being identified as exceedingly active areas for research with 11%, 11% and 8.7%,
190 respectively.



191 Figure 3: Number of articles published for the top ten subject areas involving ultrafiltration (as identified
 192 by Scopus).

193

194 These broad applications of UF are reflected by the journals in which the articles were pub-
 195 lished. A closer look on top 10 journals publishing articles on UF discloses that both sources have
 196 identified almost same journals but at a different sequence, see Table 1- A (Supplementary file).
 197 The Journal of Membrane Science was the most active in publishing on UF for both Scopus and
 198 Science Direct platforms with 854 and 814 articles, accounting for 23.8% and 33.5% of the total
 199 in the 10-year period, respectively. For Scopus, Desalination and water treatment were the next
 200 most active with 683 articles followed by Desalination with 577, Separation and Purification (372),
 201 and Water Research (274) articles. In all, the top 10 journals published 24% of the total number of
 202 articles, with the remaining 76% of articles in journals each contributing less than 0.67% of the
 203 total. Whereas the contribution was 30.5% for the first 10 journals in ScienceDirect. In this context,
 204 among identified 14,882 articles in the period of interest, there were a number of the state of art
 205 reviews. The most ten cited titles and the number of their citations are listed in Table 1 below
 206 whereas Table 1- B (Supplementary file) elucidated the most productive institutions contributed
 207 to the publication's size of UF membranes research. These institutions participated in nearly 11.6%
 208 out of the total publications about UF research.

210 Table 1: Highly cited reviews from 2009-2018, as identified by Scopus

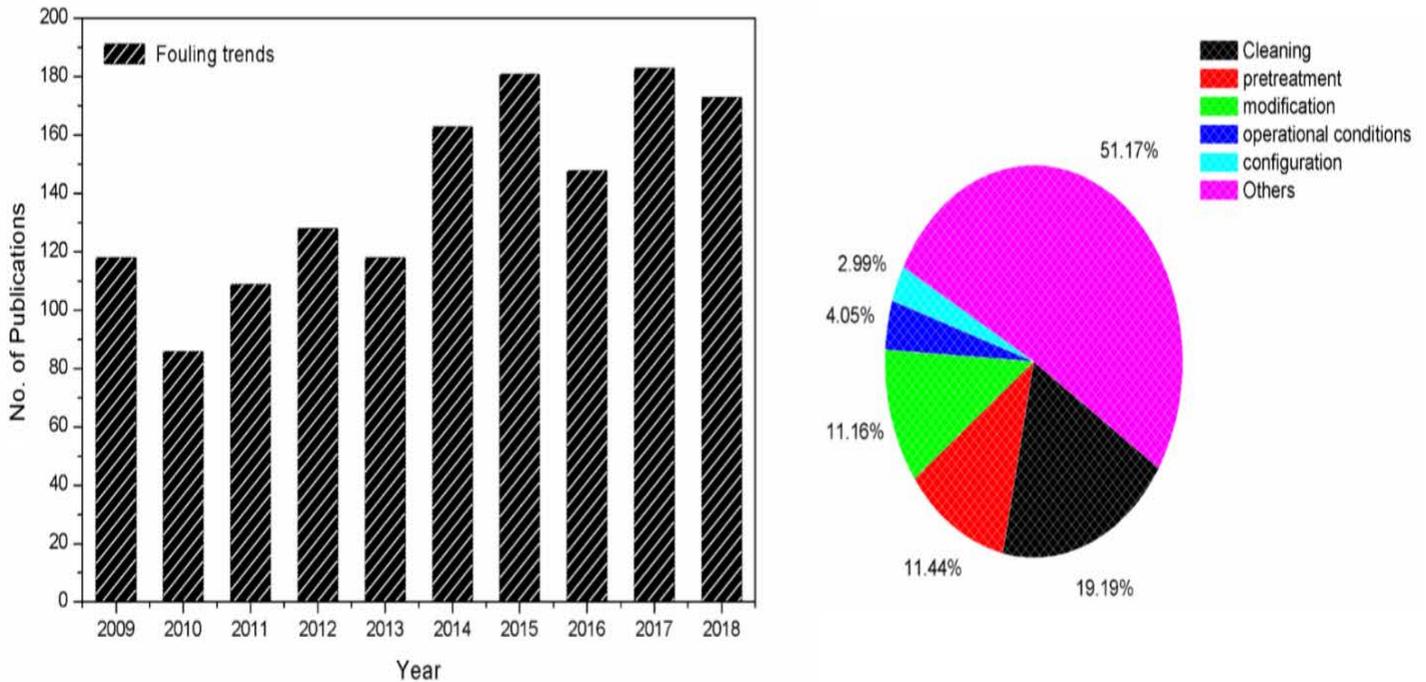
	Title of the review	Cited by	Ref.
1	Membrane fouling control in ultrafiltration technology for drinking water production: A review	414	[24]
2	Fouling of reverse osmosis and ultrafiltration membranes: A critical review	316	[25]
3	Fouling and cleaning of ultrafiltration membranes: A review	241	[4]
4	Humic substances fouling in ultrafiltration processes	137	[26]
5	Separation of functional macromolecules and micromolecules: From ultrafiltration to the border of nanofiltration	99	[10]
6	Ultrafiltration in Food Processing Industry: Review on Application, Membrane Fouling, and Fouling Control	98	[19]
7	Role of electrostatic interactions during protein ultrafiltration	55	[27]
8	Recycling of poultry process wastewater by ultrafiltration	49	[28]
9	Metal removal from aqueous media by polymer-assisted ultrafiltration with chitosan	40	[29]
10	Removal of heavy metals from wastewater using micellar enhanced ultrafiltration technique: A review	38	[30]

211

212

213 **4 Prevailing research trends (2009-2018)**

214 Among many topics, fouling of UF membranes has been an area of significant interest in the last
215 10 years. It was the single largest area of research interest detected in this study, as mentioned in
216 the previous section. Undoubtedly, membrane fouling presented a serious obstacle restraining the
217 capability of ultrafiltration that could cause a higher operating cost due to increased energy de-
218 mand, more labour for maintenance, cleaning chemical costs, and shortening membrane lifespan.
219 Therefore, and unsurprisingly, calls for effective and efficient methods for its control and minimi-
220 sation has no approaching terminus on the horizon [4,31,32]. Figure 4, manifest the number of
221 published articles on this topic each year from 2009 to 2018. About 85-186 papers were published
222 annually, totalling to 1407 in the course of last decade. Apparently, this statistic review paper has
223 identified three periods of year-on-year growth from 2010-12, 2013-15, and 2016-17. The trend in
224 research on fouling is on an upward trajectory overall, from peaks of 118 papers in 2009, 128 in
225 2012, 181 in 2015, and 183 in 2017. Thus, the need for persistent expansion in this area can be
226 noticeably seen.



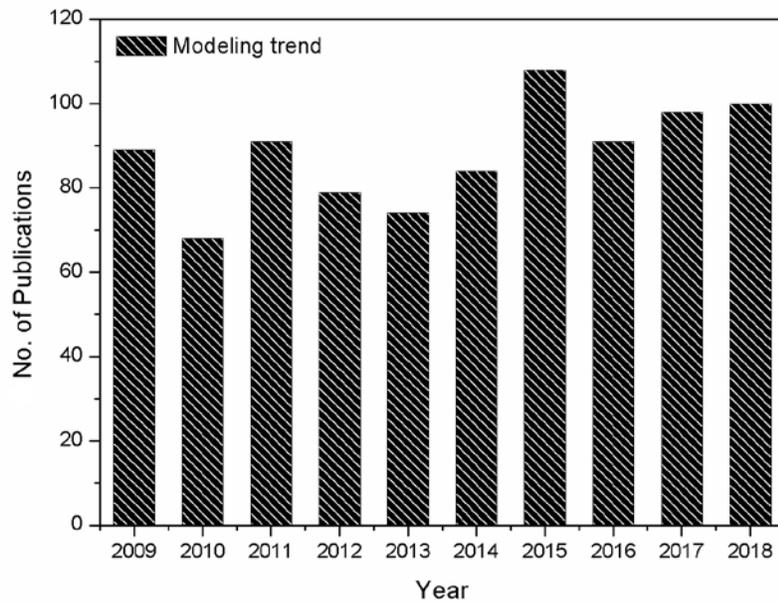
227 Figure 4: Research trend (2009-2018) on UF membrane fouling (Left), and main research themes on foul-
 228 ing (Right), according to ScienceDirect.

229

230 A number of intercorrelated factors influences membrane fouling mechanism; process config-
 231 uration, cleaning strategies, membrane types, material properties, operating conditions, and feed
 232 solution characteristics, are some among these methods. In spite of this, membrane cleaning is
 233 presently unavoidable and an essential part of membrane filtration in industry. Cleaning has to be
 234 frequently incorporated during operation in order to minimise the permanent fouling and re-estab-
 235 lish the efficiency [4,33]. Therefore, the majority of publications (about 20%) was dealing with
 236 multifaced membrane cleaning protocols [34–36]. Following this trend, the combination of re-
 237 search publications on pretreatment and surface modification was more than 22% while research
 238 about other factors such as; operational conditions and process configurations have manifested a
 239 lower attractive research trend (Figure 4, Right).

240 Mathematical modelling in membrane operations for water treatment could be crucial to endow
 241 useful data for the plant design and salutary prediction for the performance of the membrane water
 242 treatment plant [37–40]. Variation of membrane processes (microfiltration, ultrafiltration, nanofil-
 243 tration and reverse osmosis) necessitate dissimilar prediction models due to unlike transport mech-
 244 anisms. With an appropriately reliable prediction model, operational membrane issues can be iden-
 245 tified in advance. Hence, preventive actions can be taken to mitigate their impacts on long-term

246 performance [41,42]. However, it still causes a struggle to fully understand the relationship be-
247 tween the many factors that affect the process. Therefore, modelling of ultrafiltration processes
248 was the second largest area of research with a continued popular area of research. The annual
249 number of published studies fluctuating around 88 from 2009 to 2018 (Figure 5). There appears to
250 be an uptick in the past 4 years with the maximum annual number of papers released in 2015 at
251 108, and in excess of 100 papers published in 2018. In particular, these articles are concentrating
252 on modelling solutes separation, fouling parameters, flow, and the influence of operational condi-
253 tions...etc [43–47].



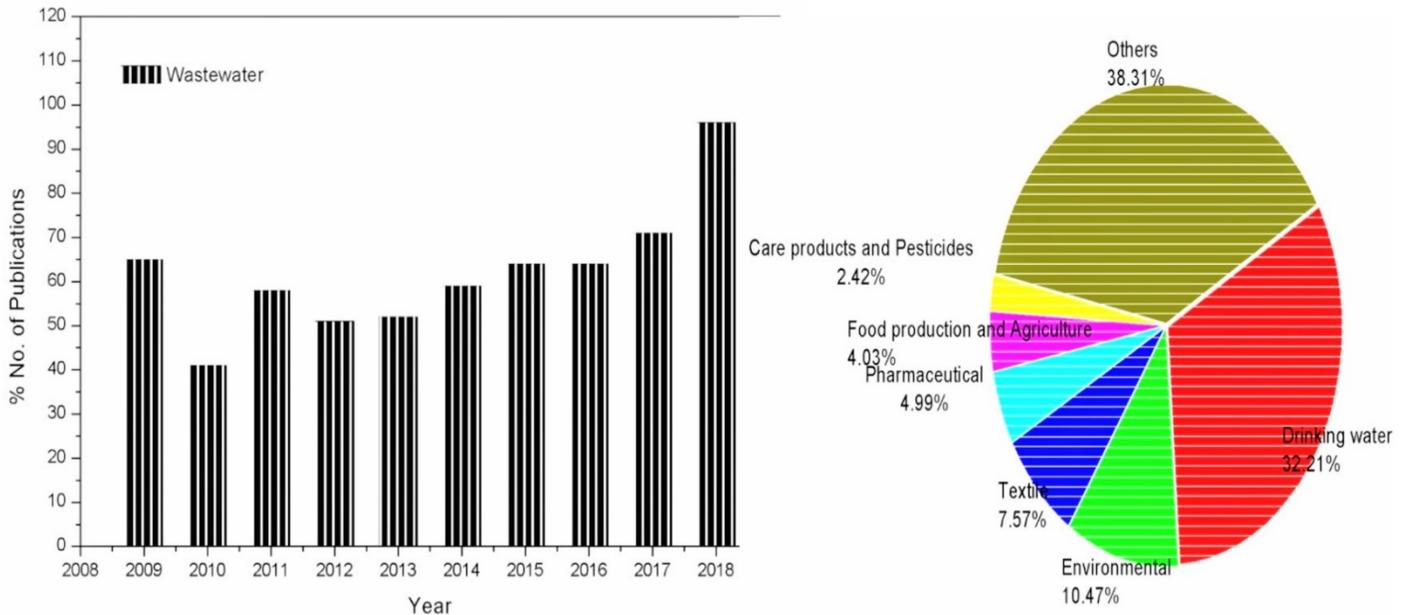
254 Figure 5: Research trend on ultrafiltration modelling (2009-2018), according to ScienceDirect.

255

256 More than 1.1 billion people on the planet do not have access to safe drinking water, while
257 another 2.6 billion people had no proper sanitation [48]. A mounting population, an escalation in
258 the requirement for resources, climate change, and pollution of accessible water resources are all
259 applying exceptional difficulties on freshwater supplies around the world [49]. Clean water defi-
260 ciencies will ultimately constrain economic growth and food supplies, while a shortage of water
261 sanitation is coupled with appalling amounts of infant mortality in the developing world. With
262 existing safe drinking water sources over-distributed, numerous regions of the world, are aiming
263 to unconventional supplies such as seawater, brackish ground and surface water, and reclaimed
264 wastewater. This explains why the third largest area of interest was dedicated to water processing.

265 Due to that, there is a steadily growing research interest related to ultrafiltration technology in
266 wastewater and desalination applications. Unsurprisingly, this is associated with the development
267 in the modern world where pollution prevention and control are a key objective of governments

268 and is forcing industry and academia. Excluding 2009 and 2018, wastewater research on UF has
 269 manifested a gradual growth across the 10 year period, increasing from 41 publications in 2010 up
 270 to 59 in 2012, barring small spikes in 2015 and 2016, then 71 articles in 2017 (Figure 6, Left). 2018
 271 was a particularly strong year for wastewater research with a 35% increase in publications on the
 272 previous year, although this stands as an outlier from the general trend. Similarly, 2009 has wit-
 273 nessed more than 58% increase comparing to the subsequent year.



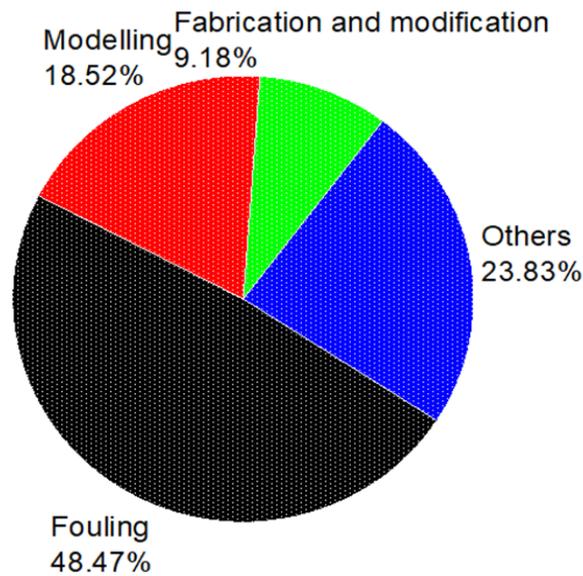
274 Figure 6: Research trend on wastewater applications from 2009-2018 (Left), application of UF for various
 275 wastewater streams (Right), according to ScienceDirect.

276

277 Over the past several decades, the discharge of pollutants into the environment has greatly in-
 278 creased due to the swift industrial expansion and fast population growth. The water pollution war-
 279 rants exceptional consideration, seeing as water quality correlates directly with the health of hu-
 280 mans and wildlife [50]. These facts have been translated into phenomenal efforts to overcome the
 281 water scarcity and quality issues, where more than 42% of wastewater applications research were
 282 dedicated to drinking water and environment. Amongst other contaminants in water, dyes from
 283 textile, paper, printing, and food industries have become a major threat to water security [51,52].
 284 Textile wastewater is one of the most difficult waste streams in the industry to treat. The process
 285 consists of several unit operations such as dyeing, desizing, printing, sieving scouring, washing,
 286 bleaching, mercerizing, rinsing, carbonization, finishing and dyeing processes [53]. According to
 287 the China Environment Statistical Yearbook in 2015, the discharge of textile dyeing wastewater
 288 was about 1.84 billion tons annually in China, posing a serious danger to the aquatic ecosystem if
 289 not appropriately treated [54]. Food industries are commonly thought as the largest source of

290 strong wastewater production which is categorised by high biological oxygen demand (BOD) and
 291 chemical oxygen demand (COD). Within the food industry, the dairy sector has the highest pollu-
 292 tion in terms of water intake and characteristics of generated effluent [55]. Numerous recent studies
 293 have determined that emerging contaminants such as endocrine-disrupting compounds (EDCs),
 294 pesticides, disinfection by-products (DBPs), pharmaceutically active compounds (PhACs), and
 295 personal care products (PCPs) are found at trace concentrations in surface waters and the toxicity
 296 of many of these compounds can possibly develop harmful human, animal and ecological prob-
 297 lems [56,57]. A quick breakdown to research trends, illustrated in Figure 6 (Right), shows that
 298 research into treating wastewater using UF membranes to produce drinking water is the largest
 299 researched sector (32%), followed by environmental (10%), with the textile (8%) and pharmaceu-
 300 ticals (5%) sectors following behind.

301 As mentioned earlier, fouling remains a major barrier limiting UF applications in treating vari-
 302 ous waters [58,59]. Figure 7 compares how fouling is a critical topic in wastewater research com-
 303 pared to other themes. According to the ScienceDirect platform, almost half of the wastewater
 304 related articles were dealing with fouling studies. The remaining half was discussing topics like
 305 modelling (18.5%), membrane fabrication and modification (9.18%), and other topics (23.83%).
 306 Table 2 below listed the top-10 cited articles concerning fouling and wastewater topics.



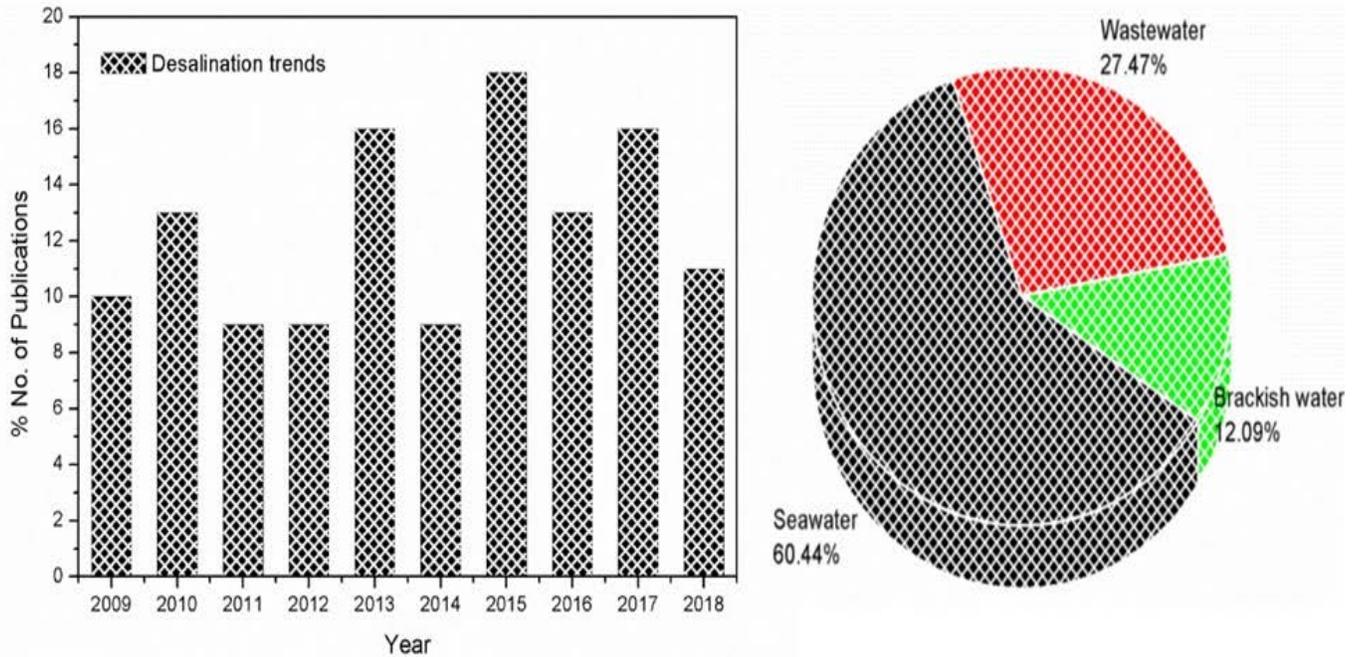
307 Figure 7: Research themes of wastewater (2009-2018), according to ScienceDirect.

308

309 Table 2: Top-10 cited articles for fouling and wastewater topics (2009-2018).

Top-10 cited articles concerning fouling topic		Top-10 cited articles concerning wastewater topic				
Title	Cited by	Ref.	Title	Cited by	Ref.	
1	Polysulfone ultrafiltration membranes impregnated with silver nanoparticles show improved biofouling resistance and virus removal	447	[69]	Treatment of micropollutants in municipal wastewater: Ozone or powdered activated carbon?	341	[78]
2	The effects of mechanical and chemical modification of TiO ₂ nanoparticles on the surface chemistry, structure and fouling performance of PES ultrafiltration membranes	359	[68]	Occurrence and removal of pharmaceuticals, caffeine and DEET in wastewater treatment plants of Beijing, China	274	[77]
3	Characteristics, performance and stability of polyethersulfone ultrafiltration membranes prepared by phase separation method using different macromolecular additives	350	[67]	Removal of synthetic textile dyes from wastewaters: A critical review on present treatment technologies	267	[76]
4	Carbon nanotube blended polyethersulfone membranes for fouling control in water treatment	310	[66]	Preparation, characterization and performance of Al ₂ O ₃ /PES membrane for wastewater filtration	184	[75]
5	Novel GO-blended PVDF ultrafiltration membranes	274	[65]	Polymer-enhanced ultrafiltration process for heavy metals removal from industrial wastewater	175	[74]
6	Improved hydrophilicity, permeability, antifouling and mechanical performance of PVDF composite ultrafiltration membranes tailored by oxidized low-dimensional carbon nanomaterials	260	[64]	Effect of additives concentration on the surface properties and performance of PVDF ultrafiltration membranes for refinery produced wastewater treatment	174	[73]
7	Organosilane-functionalized graphene oxide for enhanced antifouling and mechanical properties of polyvinylidene fluoride ultrafiltration membranes	245	[63]	Application of the Al ₂ O ₃ -PVDF nanocomposite tubular ultrafiltration (UF) membrane for oily wastewater treatment and its antifouling research	165	[72]
8	Preparation and characterization of PVDF/TiO ₂ organic-inorganic composite membranes for fouling resistance improvement	238	[62]	Permeate flux decline during UF of oily wastewater: Experimental and modeling	163	[71]
9	Preparation and properties of functionalized carbon nanotube/PSF blend ultrafiltration membranes	232	[61]	Occurrence of emerging contaminants, priority substances (2008/105/CE) and heavy metals in treated wastewater and groundwater at Depurbaix facility (Barcelona, Spain)	159	[70]
10	Synergetic effects of oxidized carbon nanotubes and graphene oxide on fouling control and anti-fouling mechanism of polyvinylidene fluoride ultrafiltration membranes	211	[60]	Tight ultrafiltration membranes for enhanced separation of dyes and Na ₂ SO ₄ during textile wastewater treatment	137	[13]

311 From the other hand, not all aspects of UF technology have witnessed an increasing trend.
 312 Herein, the desalination topic was the research area where the UF had experienced decline or stag-
 313 nation. Desalination applications are a niche within UF research especially for hybrid membrane
 314 systems [79,80], and as such have a mixed level of publications over the last 10 years (Figure 8,
 315 Left). 2015 was the most productive year, with 18 releases, whereas in 2011, 2012 and 2014 there
 316 were only 9 articles each. Other years ranged from 10-16 releases. With around 50% of the total
 317 publications, identified with the keyword desalination, were concentrated in the last four years.

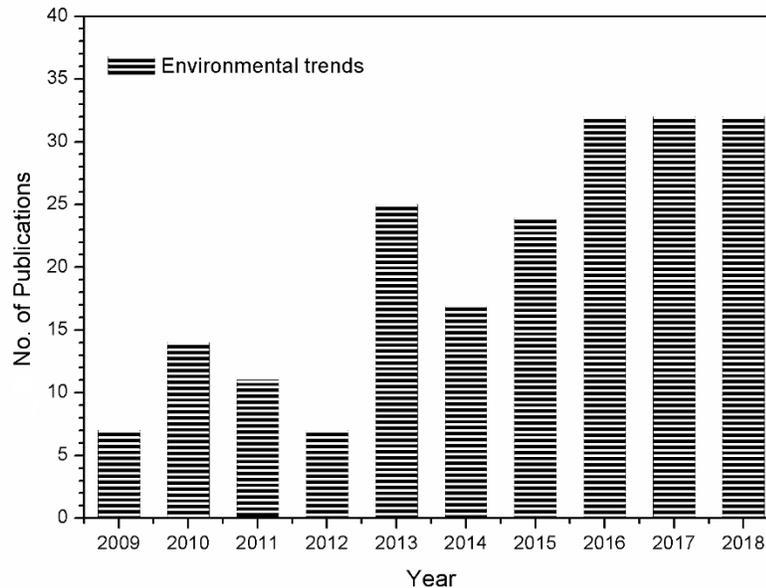


318 Figure 8: Research trend on ultrafiltration in desalination from 2009-2018 (Left), and Proportions of wa-
 319 ter feed streams desalinated utilising ultrafiltration (Right), according to ScienceDirect.

320

321 UF has been palpably employed as a pretreatment for desalination [81,82]. In difficult waters,
 322 ultrafiltration (UF) has proven to be the best technology as the final pretreatment step prior to RO.
 323 UF delivers superior water quality compared with conventional treatment due to the defined, very
 324 fine pore structure. It delivers a continuously good filtrate quality independent of feed water quality
 325 variability caused by, for instance, seasonal changes [83]. In the last decade, a number of papers
 326 have reported research into desalination utilising ultrafiltration. Desalination for water treatment
 327 was skewed between different types of feedwater, as shown in Figure 8 (Right), seawater being
 328 the most popular with 60.5% of the releases, followed by wastewater (27.5%) and lastly brackish
 329 water (12%).

330 Environmental research has also exhibited a steady level of increase over the period investi-
331 gated. The research saw a record surge in 2013 with 25 publications, more than the number from
332 the previous year 2011 and 2012 combined (Figure 9). This increase was not sustained in the sub-
333 sequent year, dropping down to 17 papers in 2014 before seeing a return to 24 papers in 2015, and
334 marked an increase of 32 releases in 2016, 2017 and 2018.



335 Figure 9: Research trend on environmental applications from 2009-2018

336

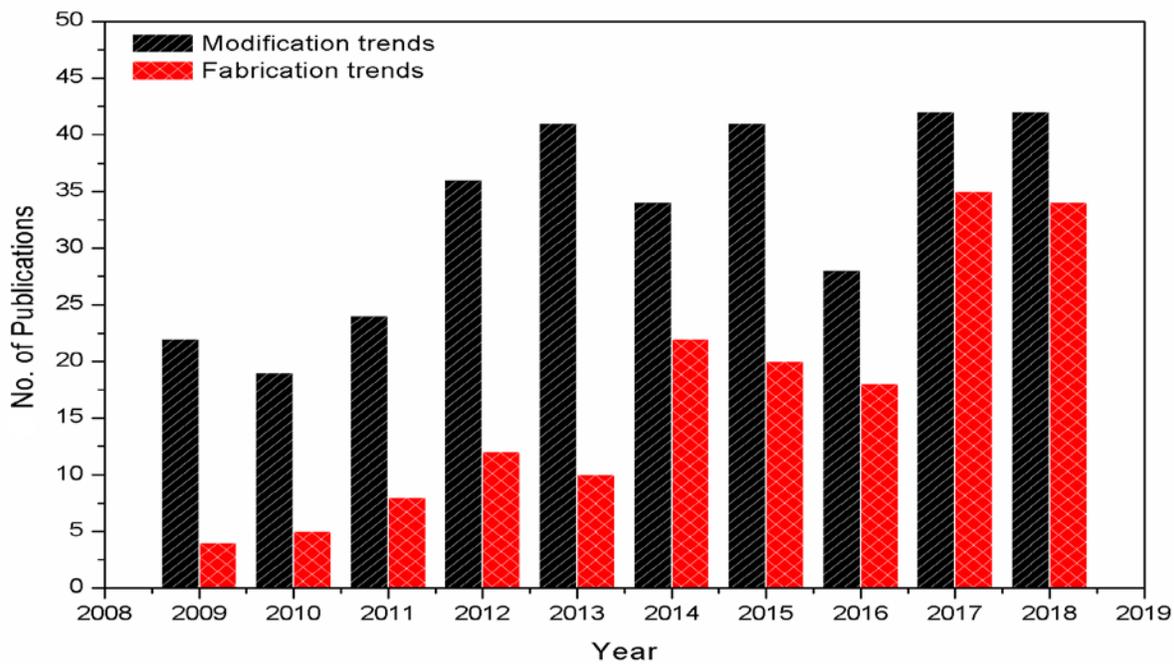
337 5 Increasing research trends (2009-2018)

338 As this review is performed, a number of research areas have disclosed a distinguished growing
339 up trends in terms of publications during the years 2009-2018. These areas consisted of UF mem-
340 brane fabrication and modification, food processing, hybrid membrane process, and design and
341 process optimization.

342 Although the total number of publications about membrane fabrication was small in 2009, a
343 general sloping uptrend declares that the technique has quite evolved over the years, see Figure
344 10. Other than 2013, 2015 and 2016, which only had 10, 20 and 18 releases respectively, and belies
345 the general trend. Publications have increased rapidly from 4 in 2009, 5 in 2010, 8 in 2011, 12 in
346 2012, then a jump to 22 papers in 2014 and 20 in 2015. The year 2017 appears to show the new
347 higher level of research interest in membrane fabrication set to continue, with 35 releases. In 2018,
348 a slight drop was witnessed compared to the previous year but still strong when compared to the
349 average values from the first 5 years in the period of observation.

350 Unsurprisingly, this evolution in the number of publications, about UF membrane fabrication
351 and subsequent modification, is expected to continue aiming to enhance the overall UF membrane

352 performance. The performance and properties of a membrane are heavily influenced by the tech-
 353 niques used for fabrication. However, phase inversion and solution wet-spinning were identified
 354 as the most common methods of UF membrane fabrication to endow with diverse membrane struc-
 355 tures, properties and performance [84]. Along with that, the materials of which the membrane is
 356 constructed can also be advantageous for the targeted application. An example of this is the appli-
 357 cation of inorganic ceramic membranes for protein fractionation [85,86]. This material has supe-
 358 rior thermal and mechanical properties over other types of commonly used membrane materials
 359 e.g. polymeric. Ceramic membranes can also be cleaned at extreme conditions, have a narrower
 360 pore size distribution allowing for more selective separation and offer the possibility for lower
 361 organic fouling [87]. Another important factor when assessing the viability of membrane material
 362 is the chemical nature of the membrane and the physiochemical environment of the solute. In this
 363 respect, pH and ionic strength determine the electrostatic interactions between the membrane and
 364 the molecules. Thus, permeation or transmission of a charged molecules through a membrane de-
 365 pends mostly on the electrostatic interactions between the molecules and the membrane and the
 366 relationship between the molecular size and the membrane pore size [88,89].



367 Figure 10: Research trend on membrane fabrication (Red), and Modification (Black) from 2009-2018.

368

369 In the meantime, research on UF membrane modification is a growing sustained area of
 370 attention with an excess of 22 papers published annually since 2009, with the overall pattern
 371 showing initial steady increase up to 41 publications in 2013, then flowed by a fluctuation period
 372 with a local maximum of 41 papers in 2013 declining to 34 in 2014 and further still to 41 and 28

373 in 2015 and 2016, respectively. Since then, there has been a steady number of papers year-on-year,
 374 when 42 papers were published during 2017 and 2018. Table 3, listed the top-10 cited articles
 375 concerning membrane fabrication and modification topic.

376 Table 3: Top-10 cited articles concerning membrane fabrication and modification topic (2009-2018).

	Title of the article	Cited by	Ref.
1	The effects of mechanical and chemical modification of TiO ₂ nanoparticles on the surface chemistry, structure and fouling performance of PES ultrafiltration membranes	359	[68]
2	Characteristics, performance and stability of polyethersulfone ultrafiltration membranes prepared by phase separation method using different macromolecular additives	350	[67]
3	Influence of polydopamine deposition conditions on pure water flux and foulant adhesion resistance of reverse osmosis, ultrafiltration, and microfiltration membranes	243	[90]
4	Synergetic effects of oxidized carbon nanotubes and graphene oxide on fouling control and anti-fouling mechanism of polyvinylidene fluoride ultrafiltration membranes	211	[60]
5	A bioinspired fouling-resistant surface modification for water purification membranes	206	[91]
6	Effect of graphene oxide concentration on the morphologies and antifouling properties of PVDF ultrafiltration membranes	198	[92]
7	Sulfobetaine-grafted poly(vinylidene fluoride) ultrafiltration membranes exhibit excellent antifouling property	197	[93]
8	Podocyte-secreted angiopoietin-like-4 mediates proteinuria in glucocorticoid-sensitive nephrotic syndrome	196	[94]
9	Preparation and characterization of poly(vinylidene fluoride) (PVDF) based ultrafiltration membranes using nano γ -Al ₂ O ₃	192	[95]
10	Highly hydrophilic polyvinylidene fluoride (PVDF) ultrafiltration membranes via postfabrication grafting of surface-tailored silica nanoparticles	185	[96]

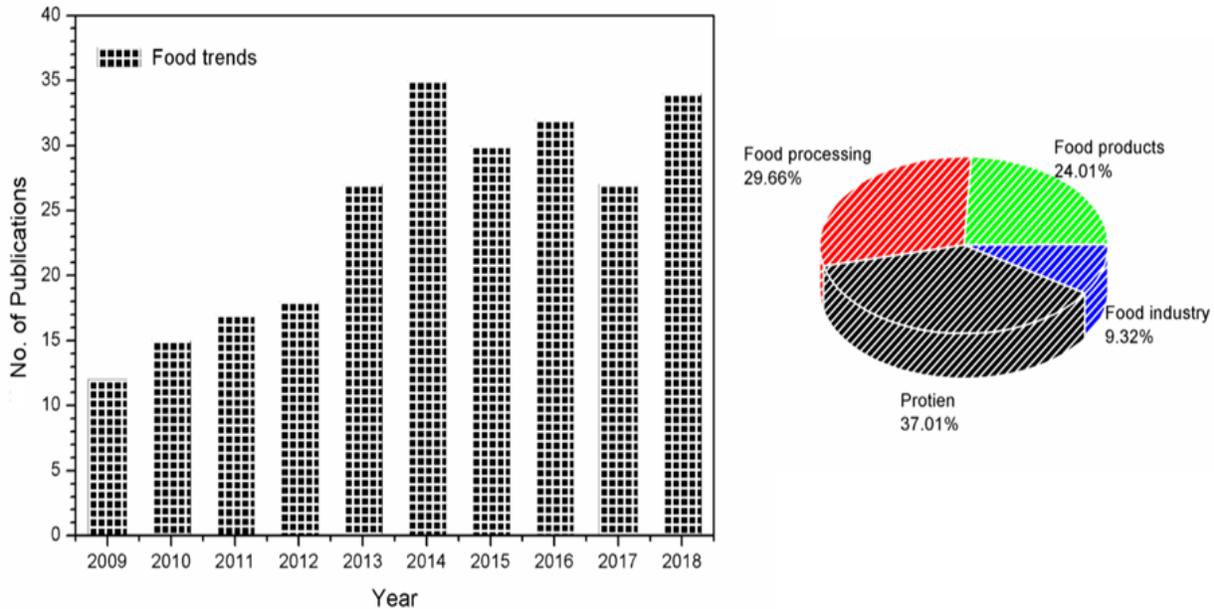
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378

379 Membrane filtration, as a separation technique in the food industry, is becoming increasingly
 380 popular compared with other conventional methods such as affinity separation, chromatography
 381 and electrophoresis. While these methods have been successful in product purification, they are
 382 not suitable for large scale production due to their low throughput and high cost. On the other
 383 hand, membranes have become a powerful tool for the recovery and purification of biomolecules
 384 in large scale production. More specifically, ultrafiltration has become the most reliable technique
 385 in the fractionation of specific compounds that are needed by the food, pharmaceutical and cos-
 386 metic industries [97,98]. Similar to research trend on UF modification, food applications for UF

387 have shown a strong increase for 2014 with 35 publications, about a threefold increase on the 12
388 papers published in 2009 (

389 Figure 11, Left). The overall trend is one of growth, but this is not maintained year-on-year,
390 with the fluctuation period from 2015 to 2018, seeing 30 papers published in 2015, 32 in 2016, 27
391 in 2017 and 34 in 2018.



392 Figure 11: Research trend on food applications from 2009-2018 (Left), and breakdown of the food studies
393 paper as a function of wide topics (Right).

394

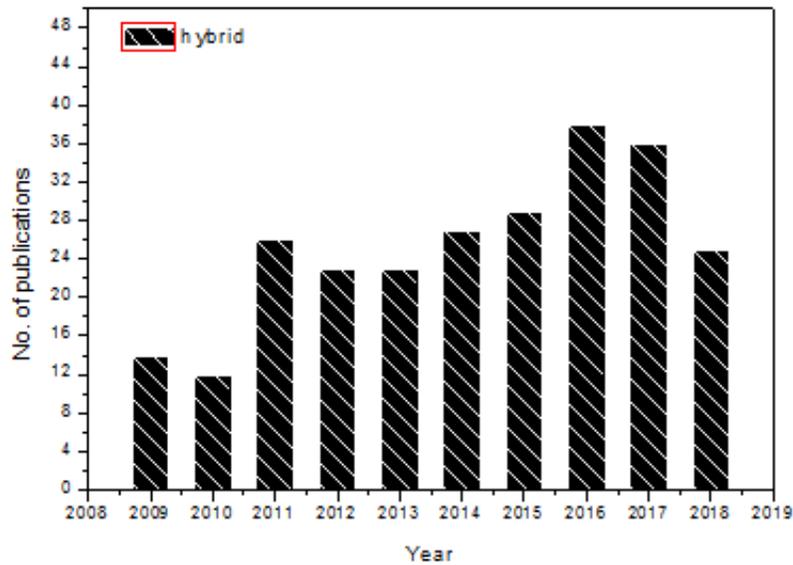
395 A further breakdown for the food analysis into the most common topics, it was identified that
396 protein, food processing, handling, industry and products were the widest areas of interest with the
397 protein section leading in research. Some research papers overlapped their focuses on these topics
398 (

399 Figure 11, Right). Protein purification was characterized as the most important application in
400 terms of ultrafiltration in the food industry (37%), followed by food processing (29.7%) and food
401 products (24%). Food industry contains the least share (9.3%) of research performed in the last
402 decade as seen, and this happens because the focus in the purification by a membrane lies as a pre-
403 treatment before other mechanisms take place, such as reactions, other purification stages, separa-
404 tion, etc. Based on the versatility acquired by the UF membrane processes over the years and their
405 wide range of applications for the marketable food products, a four simple classification can be
406 highlighted namely; dairy industry, beverage industry and fish and poultry industry [19]. These

407 three topics contributed to about 59.5% of the total number of articles within the adopted period
408 of the research.

409 This special interest paid for applications of UF in the food industry, as measured by the size
410 of publications, reflects the superior role of the UF technology. For instance; the properties of
411 biomolecules make them very sensitive to changes in temperature, pressure and with the addition
412 of additives that could change their chemical structure. Ultrafiltration membranes can be operated
413 isothermally and do not use any additives while pressure is controllable [88]. This compared with
414 other membranes of smaller sizes such as nanofiltration which require a higher driving force (pres-
415 sure) for the initial flux and this, in turn, would need a correspondingly high energy throughput.
416 Examples of biomolecules that can be isolated from the ultrafiltration technique in recently pub-
417 lished research are lycopene a carotenoid that gives certain fruits and vegetables their red colour.
418 This nutrient has been found to reduce the risk of cancers and heart disease making it a functional
419 food and a desirable product to extract [99]. Another biomolecule of use is palm oil due to its high
420 nutritional value and application in cosmetics. It is one of the fastest-growing industries in the
421 world; however, extraction of palm oil has resulted in a brown effluent which is known as palm
422 oil mill effluent. This effluent which is mainly entered through water bodies by eutrophication is
423 enriched with organic matter. Ultrafiltration can be employed to reclaim clean water from this
424 affluent and potentially be a solution to the global water crisis [100].

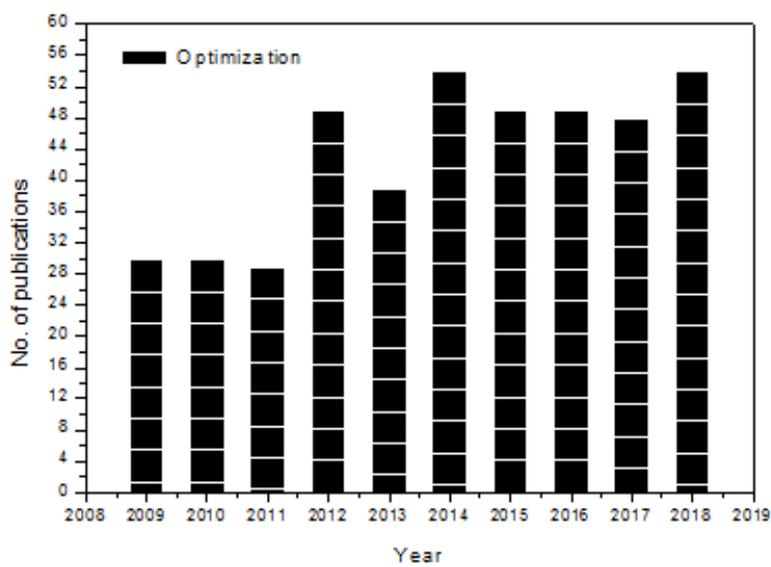
425 The concept of hybrid membrane processes has come a long way since the first inception. The
426 term hybrid or integrated membrane process refers to the integration of one or more membrane
427 processes with or without conventional unit operations to increase performance depending on the
428 type of feed and product quality required; with the main goal of these systems to increase the purity
429 of the product [101]. Hybrid membrane systems are also efficient in reducing the operating costs
430 and environmental pollution and hence make the overall process more efficient. Research on ap-
431 plications of hybrid UF processes has experienced a rising level of interest, with at least a threefold
432 increase in publications from 12 in 2010 to 38 in 2016 (Figure 12). The trend of strong upward
433 growth saw 23 articles in 2012, 29 in 2015, up to a maximum of 38 in 2016. Unexpectedly, this
434 surge was followed by a continuous decline within the past two years, with 36 and 25 articles in
435 2017 and 2018, respectively.



436 Figure 12: Research trend on hybrid UF membranes and processes (2009-2018).

437

438 At most, research articles published under the Optimization of UF membrane processes and
 439 applications subsection were varied at most between enhancing operational parameters, cleaning
 440 protocols, experimental design and fabrication conditions [102–105]. Virtually, optimization of
 441 UF membrane processes has not shown a palpable surge in recent years (Figure 13). The only a
 442 step-change has occurred between 2011-14 from 29-54 papers published annually. This annual
 443 trend has continued around 51 ± 3 publications since 2014, and onwards.



444 Figure 13: Research trends in design and process optimization (2009-2018).

445

446 **6 Potential trends in UF membrane filtration**

447 Huge advances in UF membrane processes and applications have been gained over since the first
 448 modern industrial UF membrane appeared in 1960 [106]. Even though that, there is still a long
 449 way to go with respect to membrane materials and modules, technology advancements, regulatory
 450 issues, applications, cost, system capacity and standardization [107]. As expounded earlier in sec-
 451 tion 3, fouling, modelling and wastewater, are rated as the major research areas while dominated
 452 half of the total publication's size. Excluding wastewater, a closer look at the top five UF research
 453 areas (Figure 3) discloses that the main theme of articles is concentrating on enhancing overall
 454 costs and technology advancements rather than the wide spectrum of UF applications in a variety
 455 of industrial sectors. Notwithstanding these research trends, many specific research areas were
 456 expected to endow greater research outputs if compared with their existing trends. One of these
 457 potential applications, that directly correlates to human health and needs to be further addressed,
 458 is medical applications. The concept of ultrafiltration has been widely employed in the medical
 459 industry as a component in implants [108], biosensors [109,110], diagnostic assays [111]and drug
 460 delivery systems [112,113]. Versatile molecular weight cut-off UF membranes have been utilized
 461 to retain microorganisms and constituents [114,115]. Also, patients with different pathologies un-
 462 dergo treatment including ultrafiltration applications with the goal of removal of toxins from the
 463 blood [116]. However, as can be seen in Figure 14A, there is no trend occurring in the medical
 464 aspect when focusing on ultrafiltration membranes. If 2012 was not taken into account, there
 465 would be only two or three papers published annually. One possible reason for this unreliability in
 466 paper research could be due to the irregularity in the ultrafiltration technique being applied to the

467 human body. Therefore, there is always a need for further research outputs to develop functional
468 UF membranes that can meet the criteria. Similarly, extensive pore size ranges of UF membranes
469 are available for use in the pharmaceutical industry. One of these applications is controlling endo-
470 toxin and pyrogen in the manufacturing of parenteral drug products to restrain adverse reactions
471 in patients [117–119]. In addition to polypeptide and enzyme concentration along with a distinctive
472 consistency and quality [120–123]. Despite the academic publications, about pharmaceutical prod-
473 ucts, did not exceed 107 review and research articles within the last decade, they revealed a clear
474 increasing trend from 4 in 2009 to 18 publications in 2014 (Figure 14B). However, a decreasing
475 trend was recorded after that and before the peak publication appears again in 2018. Research
476 activities need exceptional efforts for developing high-performance UF membranes combines high
477 permeability and maximum selectivity. Also, the reliability of bio-macromolecules fractionation
478 that often hindered by severe fouling. A common example in the pharmaceutical industry is the
479 protein separation. A workable fractionation process is only possible for proteins with significant
480 variation in their molecular weights [124]. Therefore, surface modification of these membranes
481 through enhancing electrostatic repulsions could bestow practical solutions to the selectivity is-
482 sues, especially for proteins having convergent molecular weights.

483 Another area which necessitates further attention is the application of UF technique in the textile
484 industry. Herein, UF main functions are through concentrating dye and effluent treatment for re-
485 covering valuable products and water reuse purposes in textile manufacturing processes [125].
486 Figure 14C contains the data of the textile research trend changes in the last 10 years. Even though
487 interests in the application of UF in the textile industry was almost double of that been witnessed
488 in medicine, there was somehow an erratic trend in publications rate as has been illustrated, and
489 the general trend is one of a slight increase. This could be attributed to the moderate dye retention
490 of UF membranes. This is imperfect for dye recovery where a significant quantity of dye can pass
491 through porous UF membranes, leading to a low recovery for dye during a diafiltration process
492 [126]. Notwithstanding that, tight UF membranes have demonstrated to be a stand-alone alterna-
493 tive to nanofiltration membranes to endow an effective fractionation of dye and divalent salts in
494 the direct treatment of textile wastewater with high-salinity [13,127].

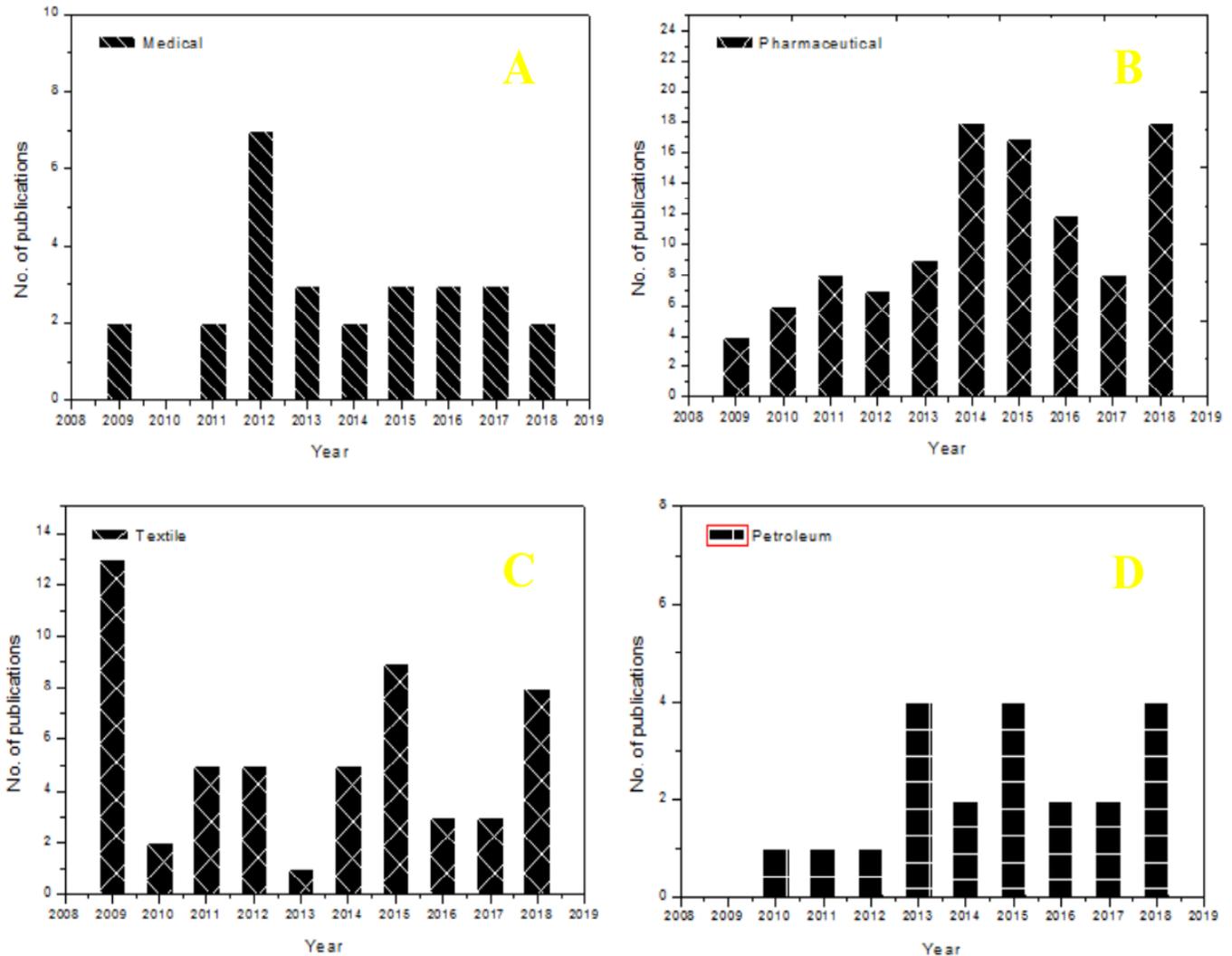
495 One of the least research fields of ultrafiltration was applied in the petroleum sector. The petro-
496 leum industry is one of the fastest-growing sectors, and it significantly participates in the economic
497 growth of developing countries [128]. Particular attention has been paid recently on the develop-
498 ment of high-performance low-cost wastewater treatment technologies [129]. Virtually, produced
499 oily wastewater composes a large volume of very complex wastewater generated from different
500 sectors. This includes; petroleum hydrocarbons, phenol, mercaptans, oil and grease, sulphide, am-
501 monia and other organic compounds that necessitates an efficient demulsification [130]. In this
502 context, low-pressure membranes (UF and MF) have demonstrated their eligibility for the reuse
503 and treatment of flowback and produced water resulted from unconventional oil and gas resources
504 e.g., shale gas and oil plays. Their share has exceeded 22% among other membrane-based tech-
505 nologies [131]. The possibility of presenting the UF/MF membranes as a compact configuration

506 stands behind their feasibility where space is very limited, which makes them ideal for on-site and
507 mobile treatment. Herein, UF/MF membranes could remove most suspended solids, turbidity,
508 grease, oil and a fraction of organic compounds. As seen in Figure 14D, no relationship in terms
509 of a number of research papers produced has been observed for each year. This happens because
510 the petroleum industry covers materials that are much larger than the possible exclusion done by
511 ultrafiltration, which only gives the opportunity for pre-treatment of materials instead of ultrafil-
512 tration working as the main method of purification. If the ultrafiltration section or the nanofiltration
513 section were to be done separately, this particular application in the industry would not have been
514 as successful, so the combination of a hybrid system could maximize the operability of the process.
515 In a recent study carried by Moslehyani et al. [132], the design and performance of a hybrid system,
516 comprising a photocatalytic reactor followed by a UF membrane cell, was evaluated against pe-
517 troleum refinery wastewater. Interesting results have been displayed due to the over 99% rejection
518 and exceptional anti-fouling characteristics.

519 Even though water processing was found as a dominant research area in UF technology here in
520 this review, further research outputs about several aspects are necessitated. One of these ap-
521 proaches is the applications of UF hybrid membrane systems in desalination plants, as an alterna-
522 tive to standardized conventional separation technologies. The felicitous operation of a seawater
523 desalination plant relies mainly on the competence of the pretreatment system utilized for con-
524 sistent production of permeate in the subsequent RO process [133]. Thus, selection between con-
525 ventional and membrane-based pretreatment is increasingly becoming tricky to make a prudent
526 decision [81]. Busch et al. claimed that about 3.4 million m³/day of UF pretreatment capacity was
527 installed in seawater RO plants in 2010. However, the recent drivers for UF technology selection
528 have been also changed where more emphasis is given to environmental aspects. This includes the
529 capability of UF to decrease chemical consumption and sludge quantities of the plants. Although
530 the greater operating cost of UF when being compared with other conventional pre-treatment is
531 still the main obstacle [134], there are many signs indicates their becoming increasingly cost-com-
532 petitive [135–137].

533 Based on the aforementioned examples above, it is can be inferred that the inevitable fouling
534 and degrading of UF membranes are the major obstacles that limit their applications, especially
535 for treating some complex feeds. This has gone hand by hand with economical considerations.
536 Fouling's level is dominated by feed and membrane characteristics, regardless of operation hydro-
537 dynamics. Membrane's material harnessed for one application may not fit another. The versatility
538 of feed compositions makes each UF membranes material and/or structure behaves differently in
539 terms of their permeability-selectivity trade-off. Thus, surface characteristics, (e.g. hydrophilicity,
540 type of functional group, charge, roughness, pore size and porosity), may endow unlike resistivity
541 to membrane's ageing and fouling in the short time. One possible solution could be through em-
542 ploying functional UF membranes targeting a specific application instead of being inclusive. For
543 instance, revising the surface characteristics of the designated membrane to enhance their selec-

544 tivity, and/or to withstand harsh cleaning chemicals or to have antibacterial characteristics. An-
 545 other promising solution, that needs to be further addressed, is through focussing on hybrid mem-
 546 brane-based processes to ensure that UF membrane avail can outweigh their cost. As a pretreat-
 547 ment step in the petroleum industry, the potential of hybrid UF membrane system should be further
 548 considered in the competition with other conventional demulsification techniques. Nevertheless,
 549 efforts are expected to continue towards the contribution of UF technology for the future of sea-
 550 water desalination plants.



551 Figure 14: Research trends of UF membrane applications from 2009-2018 for (A) Medical, (B) Pharma-
 552 ceutical, (C) Textile, and (D) Petroleum industry.

553

554 **7 Conclusions**

555 As measured by the scientific database platforms, UF membranes and processes are gaining fea-
556 tured attention inside the scientific research community. An indispensable role has been played in
557 a wide range of applications, including; food, beverage, healthcare products, bioengineering, in-
558 dustrial and municipal water, desalination and drinking water. It should be noted here that the
559 major concern was to monitor the size of research interest in UF membranes and processes over
560 the past decade. And the current statistical investigation has presented a rough estimation review
561 since many topics are overlapped with others. Excluding 2009, the number of academic papers
562 published has been steadily increased from 355 paper in 2010, 468 in 2014 to 531 in 2018, as
563 identified by the ScienceDirect platform. About 43% of the total number of publications (4547
564 articles) lay within three major scopes; chemistry, chemical engineering and environmental.
565 Among 120 detected scientific journals, the Journal of Membrane Science was the leading one,
566 comprising about 18% of the total publications. The heading topics have been discussed were
567 concentrating on fouling, modelling and wastewater, and corresponded to 27%, 17%, and 12% of
568 the total articles reviewed, respectively. To a lesser extent, other topics such as; membrane fabri-
569 cation and modification, environmental applications, food and pharmaceutical had conspicuous
570 increasing trends, particularly within the few past years. A closer look from a wide-angle enables
571 the reader to infer that the fouling was the major concern to confine the UF applications in some
572 fields, however, combining the technology with other separation tools (hybrid separation systems)
573 could endow further advantage to outweigh the technology. Based on the available statistics, there
574 is no doubt, in the recent future, that research community will persist to enhance the overall per-
575 formance of UF technology and to expand their application prospects. Ultimately, establishing
576 growth opportunities for UF membrane filtration in a wide spectrum of industries.

577

578

579 **Nomenclatures**

UF	Ultrafiltration
MWCO	Molecular weight cut-off
BOD	Biological oxygen demand.
COD	Chemical oxygen demand
EDCs	Endocrine-disrupting compounds
DBPs	Disinfection By-products
PCPs	Personal care products
PhACs	Pharmaceutically active compounds
MF	Microfiltration

580

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