Microfiltration membrane processes: a review of research trends over the past decade

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Abstract

Since its inception in the 19th century, microfiltration (MF) has evolved as a membrane-based separation technology for treating various effluents and wastewaters. This review aims to familiarize its readers with general and specific research trends on various topics in MF. The level of research interest has been measured by the number of publications in that area for each year. An increasing research trend was observed from the number of publications since 2009 to 2018 with MF as the major topic. During the past decade, MF articles have spanned in about 150 different journals, with The Journal of Membrane Science, Desalination and Separation and Purification Technology being the major ones. Major topics of interest include membrane fabrication and modification, waste water treatment and fouling studies, while a significant research increase was seen in various fabrication methods and MF application in the food sector. MF modeling still remains a topic which needs further research output, and has experienced a decline over the past years. Several potential research areas are also identified in this review which will help future researchers to materialize their efforts into the right direction.

Keywords: microfiltration, review, statistics, research trends, water treatment

Contents

1. Introduction

MF is one of the oldest pressure driven membrane applications' practiced commercially [1]. MF is capable of removing micrometer sized matter, such as suspended particles, major pathogens, large bacteria, proteins and yeast cells based on the principle of physical separation. The ability to reject a range of large scale contaminants makes MF a versatile membrane process. Figure 1 shows the removal efficiency of prominent membrane based separation processes including ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) in comparison to MF for various contaminants [2, 3]. With an increase in the molecular weight of the solute beyond 500 $g.mol^{-1}$, the separation mechanism changes from solution-diffusion (in RO) [4] to molecular filtration in which the particle size and the membrane pore diameter determines the separation characteristics [5]. MF membranes have pore diameters in the range of 0.1–5 µm. Particles with diameter greater than 0.1 µm are usually separated by a rather open membrane structure. The hydrodynamic resistance being low, such membranes require low hydrostatic pressures for a high contaminant rejection and solvent flux [6]. Nevertheless, the wide range of pore size enables MF to find applications across many areas including but not limited to pharmaceuticals [7], wastewater treatment [8], food [9], desalination [2] and biotechnology [10].

Cellulose nitrate MF membranes were first reported by Frick in 1855 [1]. The practical application of the technology was first brought about to culture microorganisms in drinking water. However, even until the mid-1960s, MF was only restricted to small-scale industrial applications. The introduction of pleated membrane cartridge in the 1970's stimulated large-scale industrial applications of MF in the pharmaceutical, water and microelectronics industry [11]. Several instances were made to modify membrane chemistry or introduce new MF membrane materials. Membrane chemistry can be further modified for improved targeted performance application. Several membrane modification methods have been recognized including, addition of various filler materials into the polymer casting solution; membrane post-treatment, crosslinking and surface modification through adsorption of external compounds. [12].MF membranes are available in both spiral-wound and flat sheet configurations. Likewise, the membrane modules and filtration units can be customized to achieve specific application goals as required. This opens up numerous possibilities for modified membrane systems for diversified applications.

Recent review papers focus on enhancing MF membrane performances by reviewing MF membrane fouling and addressing the effective challenges in applying MF technology as a pretreatment step [2] or as a stand-alone process for efficient separation processes [13-15]. Various other aspects of MF have been covered in other reviews with an emphasis on food application [16, 17]. This review aims to collate the general and specific research trends related to MF membranes and processes over the past decade, highlighting specifically those aspects of MF which have been given great research emphasis and significance. Both ScienceDirect and Scopus platforms are used to gather academic papers across the continuum of journals available. This review also highlights potential applications which needs further research input for effective utilization of the MF process. In the beginning, document source and publication trend for each year has been highlighted to advise readers on potential journals relevant for MF publication. To this end, ScienceDirect has been utilized to gather the information, otherwise Scopus is used for reviewing trends on subject matter, publication distribution among countries and other general and specific research trends. Only those publications have been included in which MF is the main topic of research or discussion thus restraining the review to relevant MF publications.

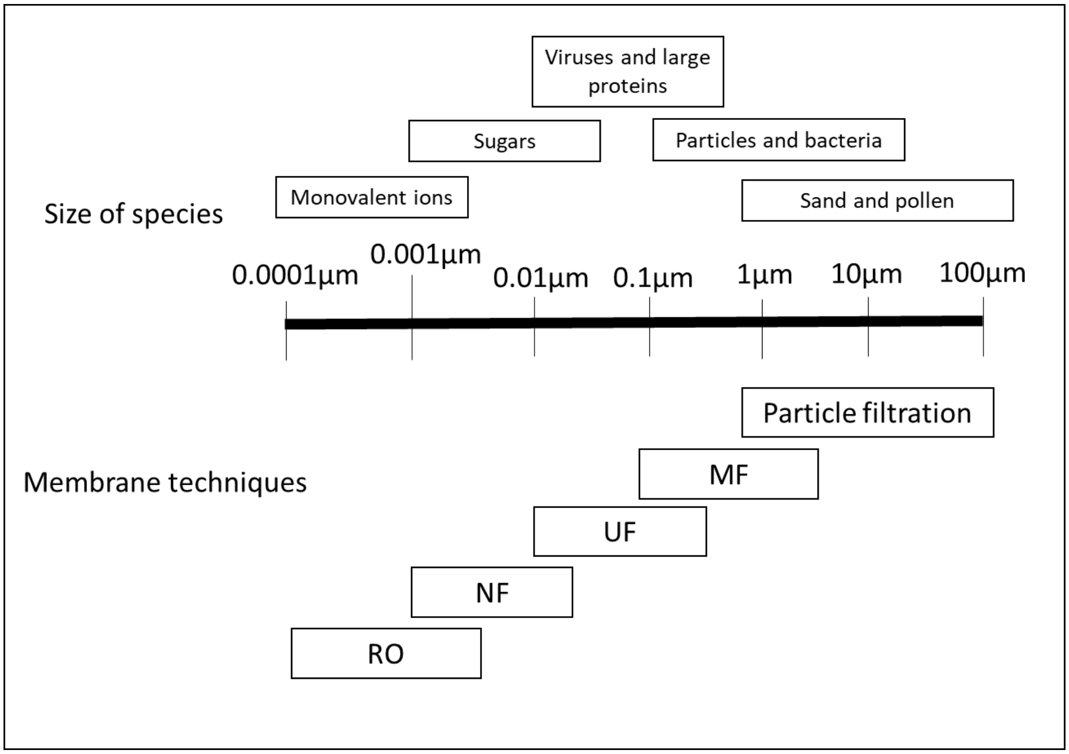

Figure 1: Prominent membrane-based processes capable of removing various contaminant sizes [2]

2. General Research Trends Since 2009

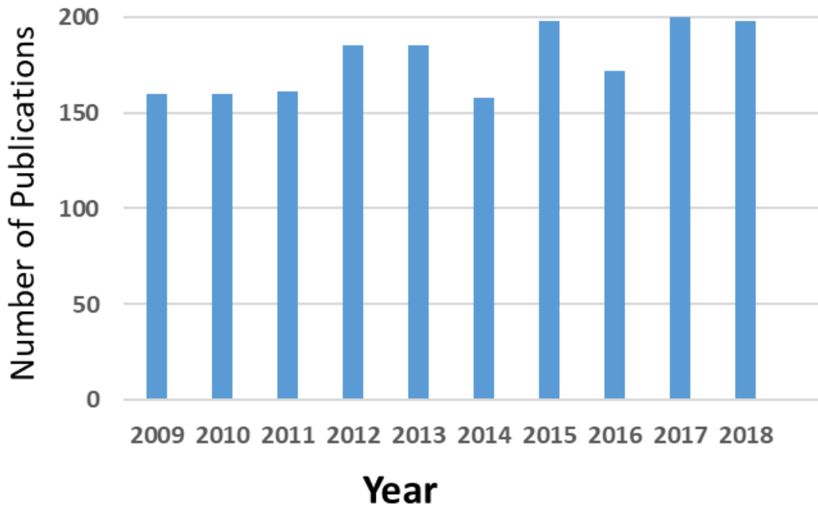

Figure 2: Number of MF papers published since 2009 according to ScienceDirect.

A general increasing trend in MF research can be observed as illustrated in Figure 2. Since 2009, a total of 1777 articles were published on ScienceDirect. These published articles include research papers, review papers, conference papers and book chapters. Among these, the largest number of document type is research articles covering more than 85% of the total publication type. Whilst there is some mild variation from year to year, nevertheless, this increasing research trend places MF as a topic of great research interest among other prominent membrane-based processes. In 2009, the total number of publications on were 160. The number of publications remained almost constant till 2011, after which it increased to 185 publications per year in the years 2012 and 2013. A small dip was observed in the year 2014, after which the publications again increased in the subsequent years reaching about 198 in 2018.

The major journals publishing articles on MF have been highlighted in Figure 3. Evidently, the main journals are The Journal of Membrane Science (22.0%), Desalination (13.0%), Separation and Purification Technology (8.6%) and Water Research (5.3%). These account for nearly 50% of total publications while the rest spread in over 150 different journals. Figure 4 shows the percentage breakdown of publications on MF across different countries during the past decade. The largest number of papers were published by China (22.9%) followed by the United States (15.1%). These two countries published more than twice as much as papers by any other country which were in the top 10. Iran and France published similar number of papers during the last decade. More than 75 different countries have been mentioned in Scopus for publishing research on MF. This clearly indicates the extent of distribution on MF research in terms of geography.

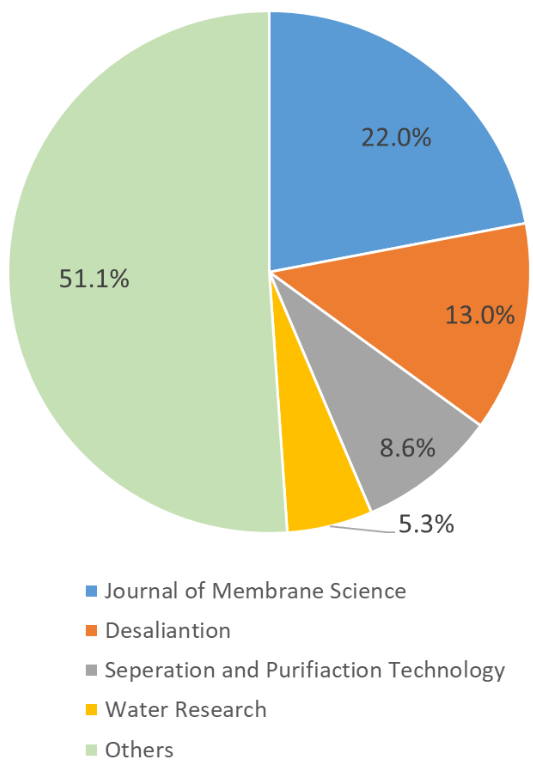

Figure 3: Major journals publishing research and review papers pertaining to MF from 2009-2018.

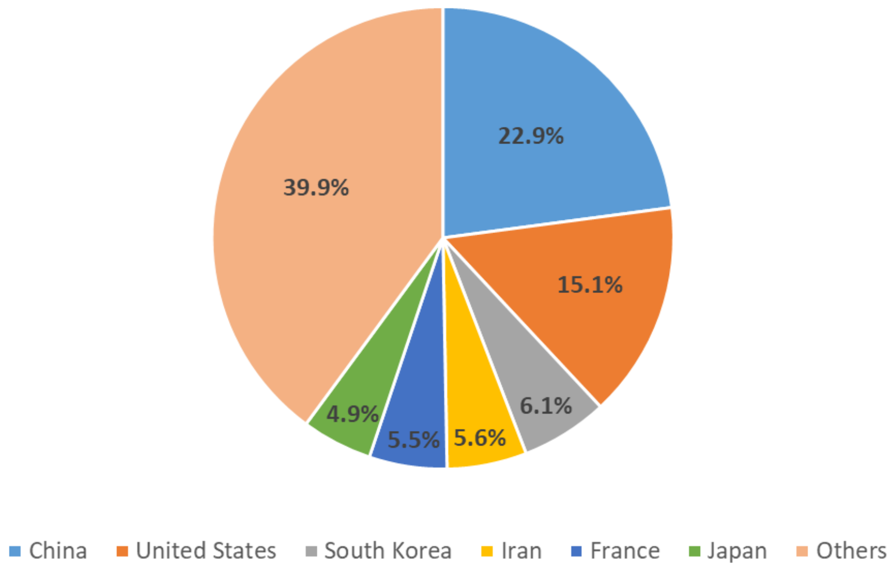

Figure 4: Distribution of the number of articles across different countries from 2009-2018.

Figure 5 represents the percentage of documents published per subject. The highest number of papers were published in the area of chemical engineering (20.6%) followed closely by the subject of chemistry. Other major areas included environmental science (14.5%), engineering (12.7%) and materials science (12.3%). From Figure 5, it is interesting to note the other diversified areas the topic of MF covers including medicine, energy, biochemistry and agriculture. These subject areas cover several research topics. Figure 6 highlights the breakdown of major topics from the total amount of publications covered in the subject areas of chemical engineering, chemistry, engineering and materials science from the year 2009 to 2018. From Figure 6, it is evident that the majority of research has dealt with water processing, and membrane fabrication and modification. Membranes constituted about 39% of total publications, while around 32% of research articles covered the topic of water processing in each of the subject areas of chemical engineering, materials science and chemistry. Other research areas include fouling (18.3%), modelling (4.5%), economics (4.4%) and review papers (1.45%). It should be noted that a publication may cover several research topics. For example, a research article on membrane fabrication may be applied for water processing and might include membrane cost analysis. Fouling is one of the biggest problems' encountered in MF membranes and thus significant research input had been made in the past decade on this topic. Its increasing research trend will be discussed in section 4. Other areas such as modeling and economics have a low research input and hence, the statistics give a clear indication to future researchers for investing more research time and effort in these areas.

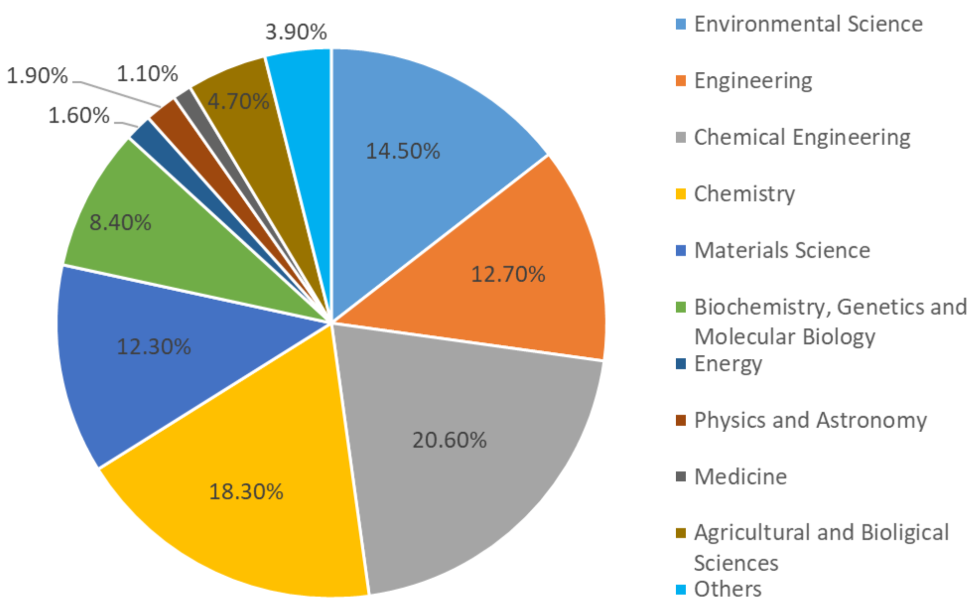

Figure 5: Percentage of documents published by subject area from 2009-2018.

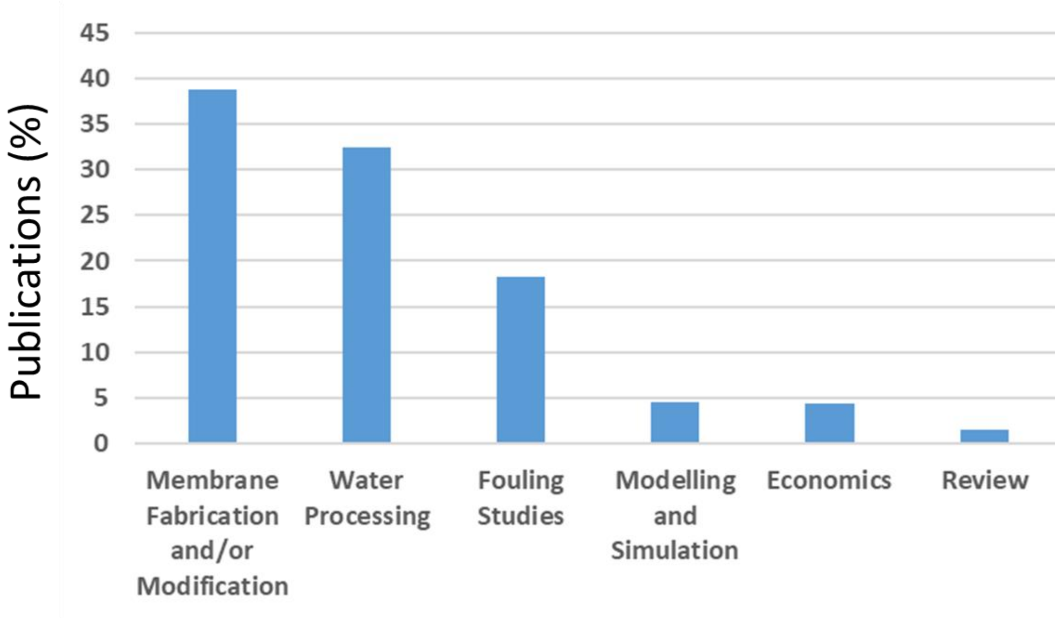

Figure 6: Research topics covered in the articles reviewed from 2009 to 2018.

MF finds potential in several applications. The four most common applications are highlighted in Figure 7, whereby the number of publications are compared for each application for each year since 2009 to 2018. In general, an increasing trend can be seen for wastewater treatment and food, while the number of publications for medicine and desalination have remained almost fairly constant over the last decade.

Water treatment is the single largest industrial application for MF. Wastewater treatment has dominated the MF market for more than a decade till present. The process is usually utilized in conjunction with other conventional or membrane based treatments. For example, Changmai et al. [18] reported an electrocoagulation-MF hybrid system for the treatment of oily wastewater. The MF process was specifically utilized to clear out the flocculants from the water during the first treatment step. It is worth stating that during the review, we came across papers which reported both desalination and wastewater treatment. Thus, some publications in Figure 7 overlap with each other in terms of application. For example, Mouiya et al. [19] reported ceramic MF membranes for both desalination and industrial wastewater treatment. Similarly, Ali et al. [20] reported the fouling behavior MF system for both water and wastewater treatment. This is followed closely by the food industry where MF membranes are utilized on a large scale for effective removal of bacteria from food products. For example, an important industrial food application is milk filtration whereby many different kinds of milk have been reported to undergo MF for the removal of spores [21] , bacteria [22] and retarding of gelatin and sediment formation in ultra-high temperature milk [23]. Owing to MF membranes' capacity to reject a variety of large scale contaminants, the desalination industry has been utilizing MF process as a pretreatment step prior to NF or RO for a considerable period of time [2]. Industrial application of desalination holds immense importance, however, its research is mainly limited due to the increasing trend of other conventional and non-conventional pretreatment methods [2]. The field of medicine is the lowest observed area of research found in this review. However, medicine has a higher research input in other membrane-based processes such as ultrafiltration where the membrane pore diameter is more appropriate for effective removal of certain contaminants [24] and hemoconcentration during cardiopulmonary bypasses [25].

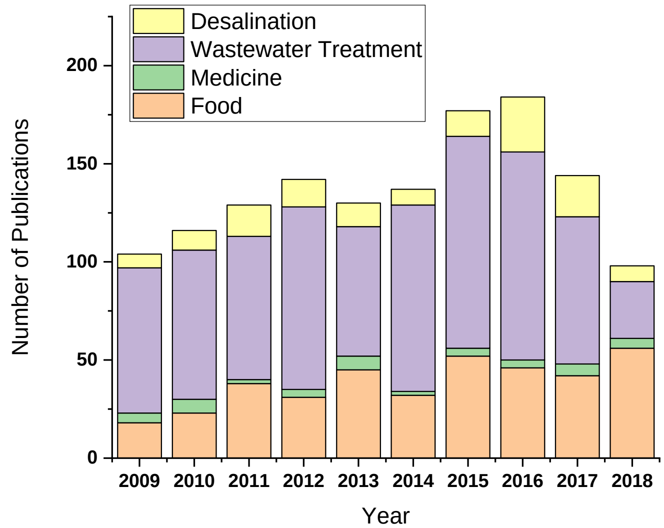

Figure 7: Total number of papers on different MF applications from 2009-2018.

3. Dominant Research Areas

As discussed in section 2, water processing forms a dominant research area for MF processes, with waste water treatment forming more than 58% of publications in terms of MF applications. Although desalination forms an integral part of water processing, it was not identified as a major research area in MF. Research on MF membranes has gained much importance and continues to do so as further developments in the field necessitate for efficient, cost-effective MF processes and thus, membrane fabrication and modification form a yet important research area in MF, as discussed in section 3.2. Lastly, membrane fouling needs special attention in this review as numerous studies have reported on fouling control for MF membranes.

3.1Waste Water Treatment

Owing to the pore size of the MF membranes, ranging from 0.1 to 1 micron, MF membranes are very popular as one of the initial filtration stages' for waste water treatment. Research trend in wastewater treatment involving MF has remained fairly high throughout the past decade, with at least about 70 publications each year except for a dip in 2018 as illustrated in Figure 8. More than 100 research papers were published in the years 2015 and 2016, after which a declining trend can be observed till 2018. It would be interesting to see the research trend in the coming years, where the MF technology still proposes several areas for improvement in wastewater treatment. Several subtopics within wastewater treatment were identified, with the prominent ones highlighted in Figure 9 including wastewaters from various heavy industries, dairy and agriculture. Dye removal was seen as the most published area totaling 144 publications over the decade. This comes without a surprise where urbanization has led to numerous textile industries and thus an immediate need for improved MF membranes for superior dye rejections. For example Tahri et al. [26] reported their work on treating a combination of dyes from actual dying cycles used in textile industries. Other research publications regarding dye removal from waste water treatment included the fabrication of novel MF membranes and modification of the existing ones. Following rapid industrialization, wastewaters from several other industries such as pharmaceuticals and petrochemicals have raised awareness on the implications of untreated disposals from these industries. Thus, increasing research is being carried out on the wastewaters disposed of from these heavy industries for safe discharge of water into the sea.

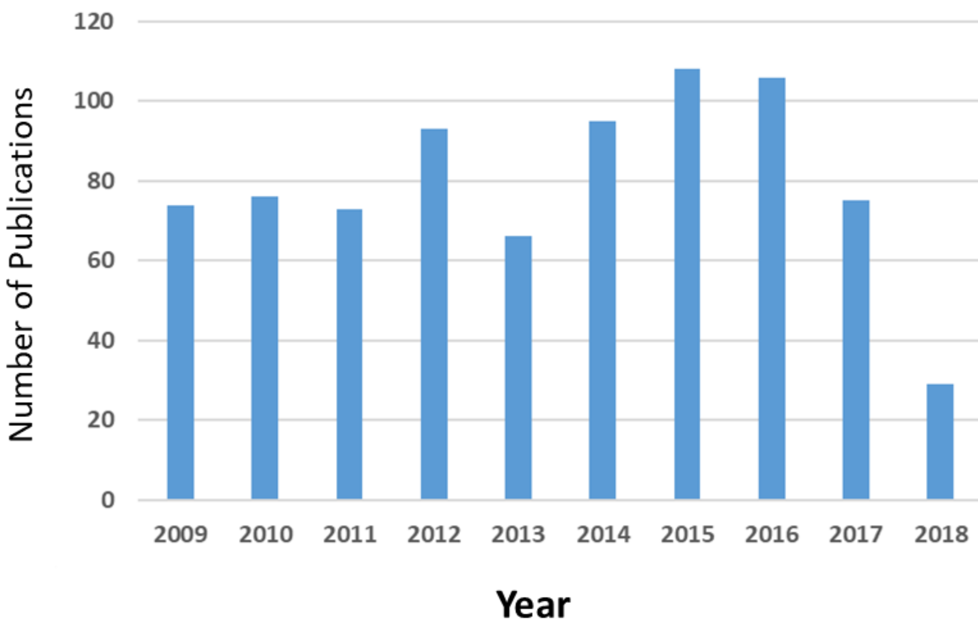

Figure 8: Research trends in wastewater applications for MF (795 total papers) from 2009-2018.

Another interesting subtopic in waste water treatment was found to be drinking water. Though NF and RO are predominantly used for producing potable, fresh water from sea water, it was identified that MF membranes are increasingly being researched to treat waste water to obtain safe drinking water. This comes without a surprise where with growing urbanization, treatment of polluted water is a key for water recycling and reuse. Several countries have become critical of water sustainability, making it a top priority in their water management policies. For example, Singapore has built wastewater plants for sewage water treatment which could serve 50% of its water demands in the near future. In 2003, its launch of 'NEWater' project provided the more cost-efficient and eco-friendly solution to water shortage problems. The process is based on a three stage purification system involving MF as an initial step, followed by reverse osmosis (RO) and ultraviolet disinfection [27], from which sewage water was treated to be declared drinkable water. Thus numerous research articles now stress on this aspect. For example, continuous MF submerged membranes are widely being researched for this purpose, with several pilot and full scale setups already established around the world [28, 29]. The heart of such processes is the MF membrane, and thus, most of the dominant research areas in MF overlap, especially with wastewater treatment and membrane fabrication and modification (section 3.2).

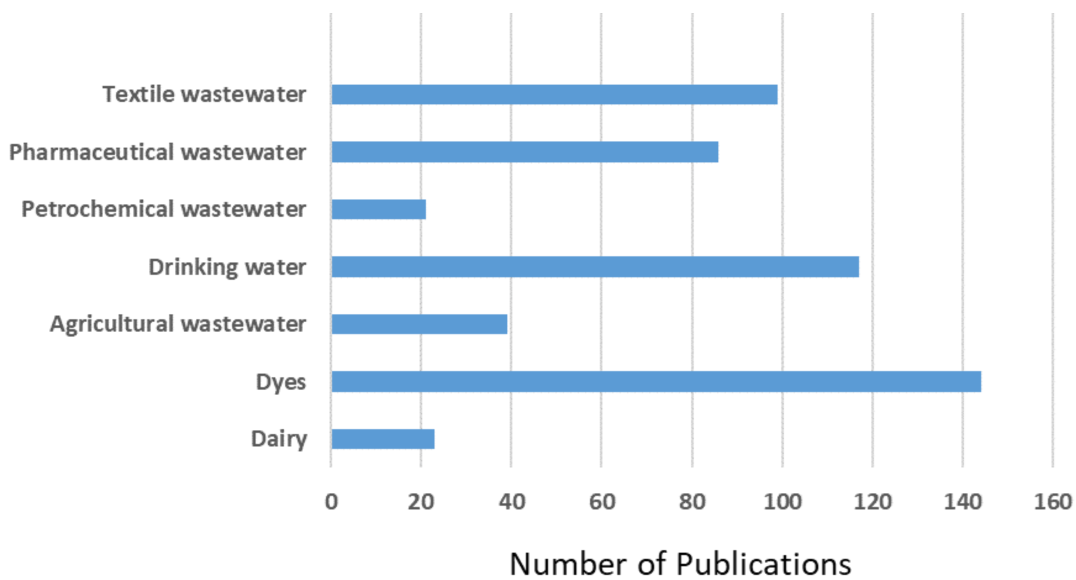

Figure 9: Subtopic areas in wastewater MF research

3.2Membrane Fabrication and Modification

Being a membrane process, research in MF is mostly concentrated in increasing membrane performance in terms of permeate permeability, selectivity, resistance to fouling and reduction in cost. These are mostly carried by the fabrication of novel membrane materials and modification to the existing ones. Early MF membranes were mostly based on cellulose nitrate. Decades of membrane development led to membrane optimization, whereby numerous researchers varied polymer concentrations, studied membrane pore sizes, varied fabrication techniques and used several novel materials [1]. Figure 10 shows the number of publications on MF membranes over the past decade, with the total number of publications totaling 1335. A fairly high research trend can be observed, with around 133 publications on an average each year. A definite increase was observed from 115 publications in 2009 to 145 publications in 2013. Around 119 papers were published in 2014, after which, again a spike was observed in 2015 with total publications of 144. The highest number were published in 2016 totaling 151. These high trends might be attributed to the International Conference on Desalination using Membrane Technology which was first introduced in 2013. Papers from this conference were then published in the journal *Desalination*, in and after the years the conference was held in 2013, 2015 and 2017. Apart from this, the need for the development of novel membranes might be linked to the surge to serve several industries including the pharmaceutical and agricultural which have put high demands on better, optimum membrane properties for high contaminant selectivity [30] .

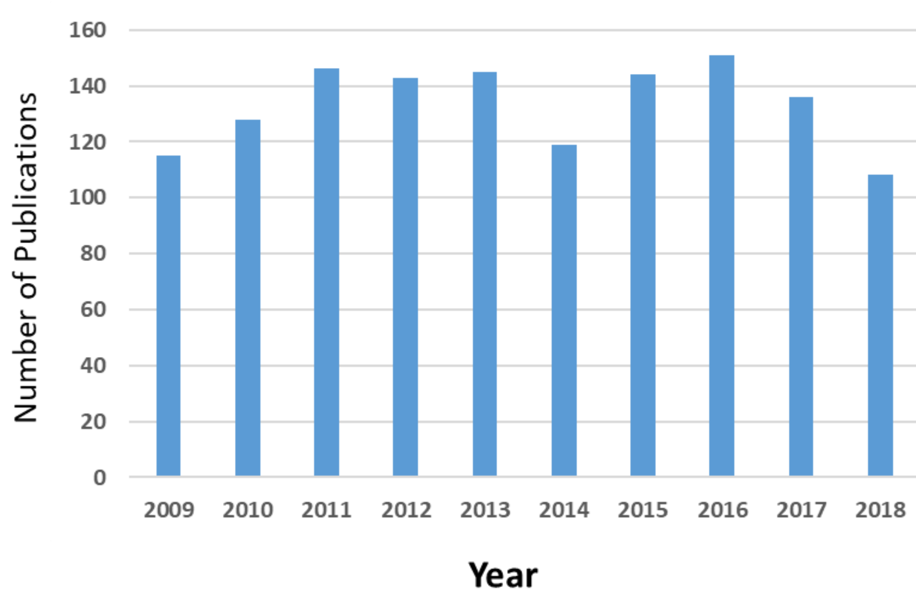

Figure 10: Research trends in membrane fabrication and modification from 2009-2018 (total papers: 1335)

Several MF membrane materials were identified during the course of 10 years. Similar to NF and RO membranes, MF membranes may also be fabricated in several layers with and without fillers, consisting of active and support layers forming the membrane structure [31]. Figure 11 shows the most common organic and non-organic materials identified in this review for MF membrane fabrication and modification in terms of percentage, while table 1 highlights progress in several important membrane materials reported during the past decade. The most popular material was poly (vinylidene fluoride) (PVDF) (16.5%). Bearing excellent chemical resistance, good membrane-forming ability and high thermal stability, PVDF has been widely used for water treatment purposes. Nevertheless, its hydrophobic nature has drawn extensive attention for membrane modification to overcome membrane fouling and increasing its hydrophilicity. Thus, many research papers have focused on various methods to improve this aspect of PVDF [32]. Other most common MF membrane materials found in this literature review included PES (10.4%), CA (7.7%), PP (7.2%), PSf (7.1%) and PA (6.1%). Figure 11 highlights the other polymeric materials. Besides polymers, several non-organic materials were identified, either as monolithic membranes [33] or mixed matrix membranes (MMM), with ceramic nanoparticles embedded within a polymer matrix [32]. SiO_2 (8.7%) and Al_2O_3 (7.8%) were the main non-organic MF membrane materials with the others shown in Figure 11. This shows the wide variety of materials available for this application and the sheer amount of work put into research for recognizing membrane properties and altering them for specific industrial use.

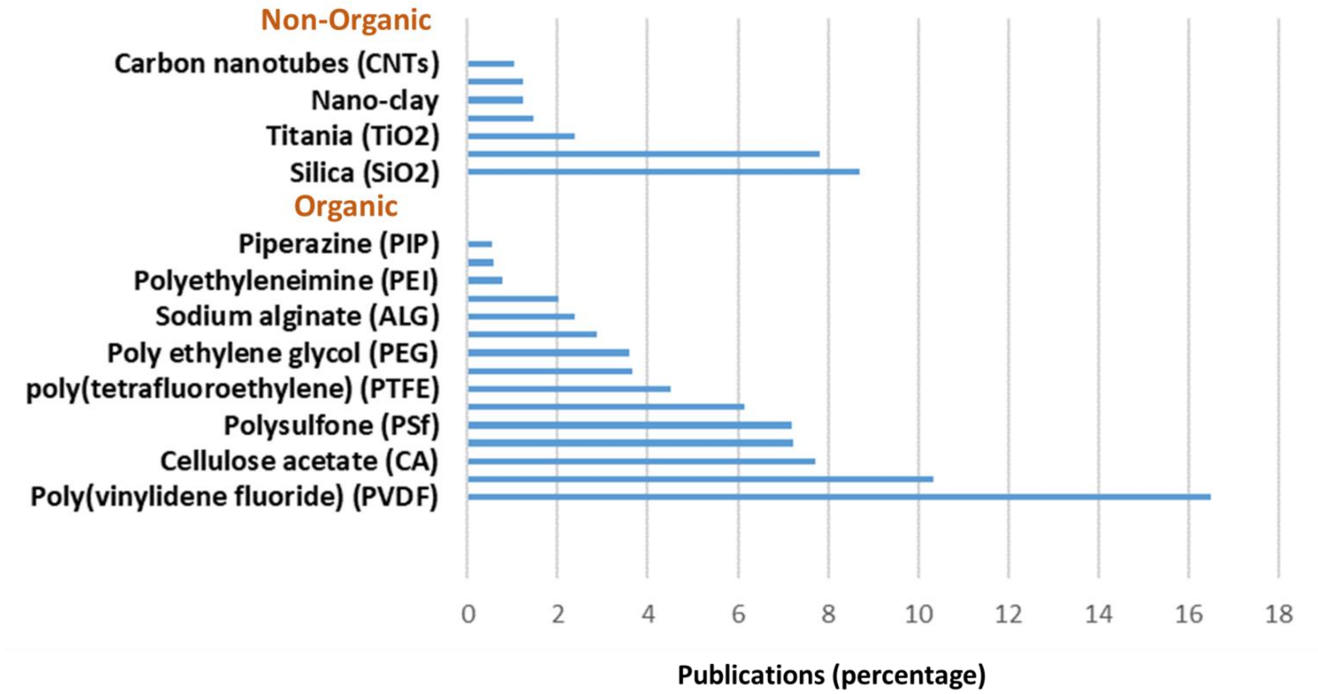

Figure 11: Major chemicals identified to fabricate and modify MF membranes found in this review

Table 1: Recent progress on MF membrane materials over the past decade

Reference	MF Membrane	Performance
Jedidi et al. [34], 2009	Mineral coal fly ash	Chemical oxygen demand and dye color removal was 75% and 90%, respectively.
Zhang et al. [35], 2009	Porous cordierite hollow fiber membrane	-
Guo and Ulbricht [36], 2010	Polypropylene microfiltration membrane modified via entrapment of an amphiphilic alkyl oligoethyleneglycolether	60 μg/cm^2 of adsorbed for bovine serum albumin (BSA) for a solution of 1 g/L
Abadi et al. [37], 2011	A tubular ceramic MF (α-Al_2O_3)	85%, 100% and 98.6% reductions of oil and grease content, TSS and turbidity, respectively. TOC removal efficiency was higher than 95% for all the experimental conditions.
Wang et al. [38], 2012	Electrospun polyacrylonitrile (PAN)/non-woven polyethylene terephthalate (PET))	Very high rejection ratio of micro-particle and bacteria (Log Reduction Value=6)
Li et al. [39], 2013	Poly (trimethylene terephthalate) nanofiber membrane	Rejection rate of 99.6% to TiO_2 suspension.

Zhong et al. [40], 2013	Asymmetric tubular α-Al_2O_3	The membranes were capable of removing ultrafine $Mg(OH)_2$ precipitates and removal rate of Mg^{2+} cations was higher than 99.9%.
Zhao et al. [41], 2014	PVDF/GO	The introduction of GO improved antifouling properties of composite membranes. A higher flux recovery ratio of 81.12%, compared to the bare PVDF membrane (52.63%).
Fischer et al. [42], 2015	TiO_2 nanoparticles on PVDF	100% of methylene blue was degraded
Yuan et al. [43], 2017	Polyamide-12 through selective laser sintering	Laser energy density of 0.1 J/mm2 results in membranes with the highest rejection and relatively high pure water flux.
Foorginezhad et al. [44], 2017	clay, zeolite and polyethylene glycol composite	95.55% removal of crystal violet at 1 bar and 90.23% removal of methylene blue at 1.5 bar transmembrane pressure.
Woo et al. [45], 2018	PTFE modified with magnetite nanoparticles	High fouling resistance with a water flux higher than 50% of initial flux for 500 ppm LUDOX SM-30 solution
Bukhari et al. [31], 2018	SiC particles over a porous SiC ceramic	-
Shi et al. [46], 2018	PEG and tannic acid	High removal ratio (>98.9%) at high flux up to 4777 L/(m^2·h) for Rhodamine B dye

3.3 Fouling

Membrane fouling forms the third largest research area in MF. Undesirable deposits of various organic and inorganic species can lead to pore blockage and hence reduce membrane permeability and selectivity. Further, this often leads to decreased membrane lifetimes. MF membranes when used a pretreatment to RO, are usually required to decrease RO membrane fouling. However, membrane fouling in MF membranes itself is a great concern, especially during treatment of wastewater where several biological contaminates can lead to biofouling. Thus, the need for continuous development in this area can be clearly seen in Figure 12 which shows the number of publications on this topic each year from 2009 to 2018. Roughly over 50 papers were published each year, totaling to 631 in the course of 10 years. Small variations may be observed, however with only slight variations. Being a serious limitation in membranes, this high trend in fouling can be predicted to continue over the coming years and thus advanced fouling resistance membranes will be a topic of great research interest for future researchers.

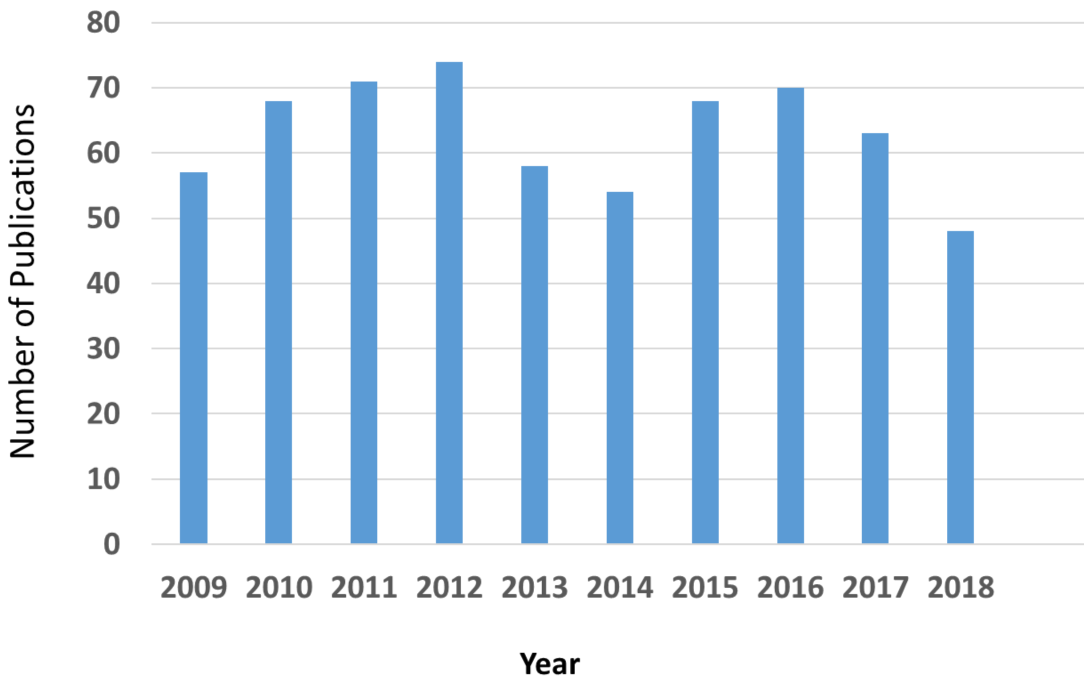

Figure 12: Research trends in membrane fouling from 2009-2018 (total papers: 631)

Majority of the fouling studies involved MF membrane modifications. Tse et al. [47] reported the grave concern of membrane fouling in membrane bio reactors (MBRs) for treating wastewaters. They developed an anti-biofouling membrane incorporated by Quorum sensing inhibiting (QSI) compounds. The compound is known to disrupt the bacterial cells. Williamson ether synthesis reaction was used for membrane synthesis consisting of QSI and a commercial polyethylene MF membrane. When treated for industrial wastewater, the modified membrane showed significant biofouling resistance compare to the unmodified one. Besides membrane modification, other publications included fouling studies for optimum MF operating conditions. Mondal and De [48] quantified the flux decline of a hollow fiber MF membrane over time by studying the growth of the fouling layer on the membrane's surface. Optimum operating conditions were identified, with limiting transmembrane pressure and Reynolds number. Besides general fouling mechanisms, efforts have also been put in understanding fouling mechanisms for specific foulants such as humic acid-protein mixtures at different solution conditions [49]. Hence, fouling studies in MF occupies a much broader perspective, covering several areas of research from membrane modification to individual fouling studies for optimized operating conditions and understanding in-depth fouling mechanisms in MF membranes.

4 Increasing Research Activity

During this review, several research areas were observed to have an increasing research trend during the years 2009 to 2018 in terms of publications. These include fabrication methods of coating, phase inversion and electrospinning techniques, while the fourth one includes food as a MF application. The phase inversion process has seen quite a noticeable research activity as shown in Figure 13. From 2009 to 2011, only about 7-10 papers were published in this area, after which the publications became almost above average till 2018. The highest numbers were reported in 2018, 29 papers in total, and thus, this research trend might be predicted to see an increasing trend in near future. The technique provides flexibility to embed numerous functional materials in the polymeric matrix for improved membrane performance [50], and thus, is the most popular method for membrane fabrication. Another fabrication method which has gained popularity is the coating method, see Figure 14. A steady increase of publications was observed on this topic over the last decade, from 22 publications in 2009 to 59 in 2018. From 2009 to 2014, almost 20 papers were published per annum (varying from 21 to 34) while at least 44 papers were published per annum between 2015 and 2018. This increasing trend is similar to the phase inversion research trend. Novel coating methods were researched during this period. For example, Qin et al. [51] reported a one-step dip coating method for coating Al_2O_3 on macroporous tubular supports while a facile sol-gel coating process was reported by Song et al. [52] for antibiofouling modification of PVDF MF membranes. Recently, there has been much interest in fabricating membranes through the electrospinning technique. The technique provides flexibility, and at the same time control over the membrane structure [53]. Figure 15 shows an evident increase in research on MF membrane fabrication through the electrospinning method. Even though the number of total publications in this area has been small, with 67 papers in total, a general increasing trend shows that the technique has quite evolved over the years. With only 1-2 papers published in each year from 2009 to 2011, a significant increase of about 12-13 papers was seen in the years 2017 and 2018. Initially, most research studies concentrated on reporting effective removal of certain substances such as humic acid [54], while recent studies concentrate more on performance parameters such as achieving high flux [55] and antifouling properties [56, 57] for improved membrane lifetimes. Owing to the versatility of the process, such properties are easier to tailor with electrospun membranes [58]. The increasing research trends in membrane fabrication techniques seems reasonable looking at the high research trends in section 3.2, whereby a continuous effort is put into modifying the existing synthesis techniques, and utilizing the present ones for novel membrane fabrication.

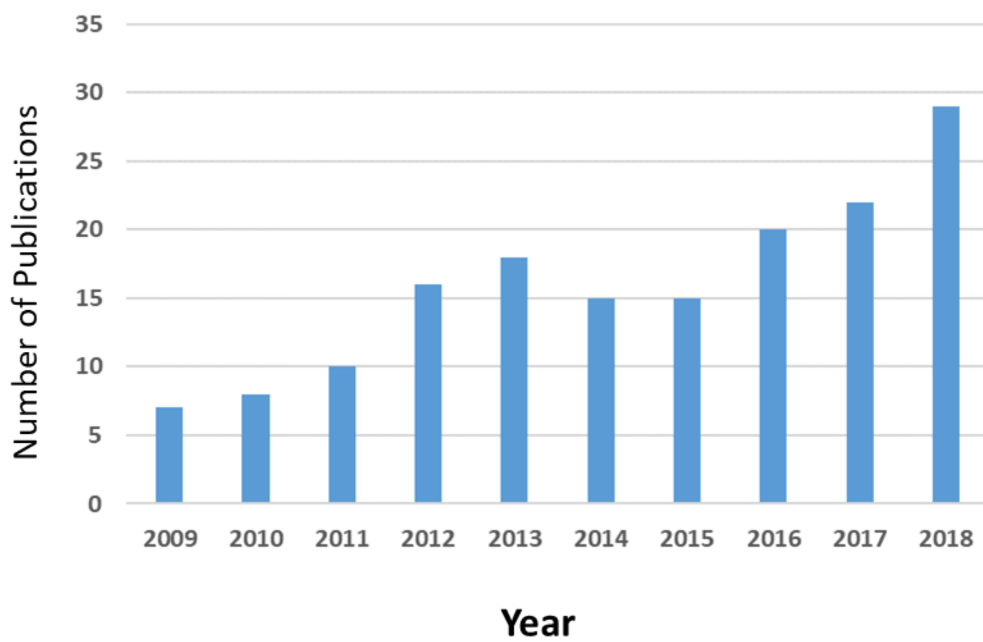

Figure 13: Research trends in phase inversion method in MF technology (total 160 papers)

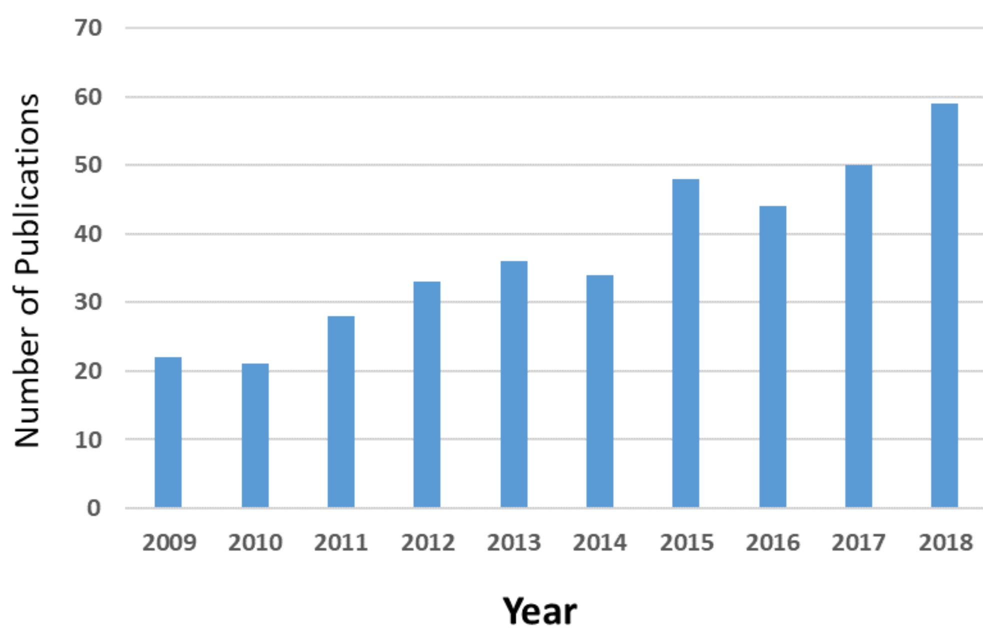

Figure 14: Research trends in coating method in MF technology (total 375 papers)

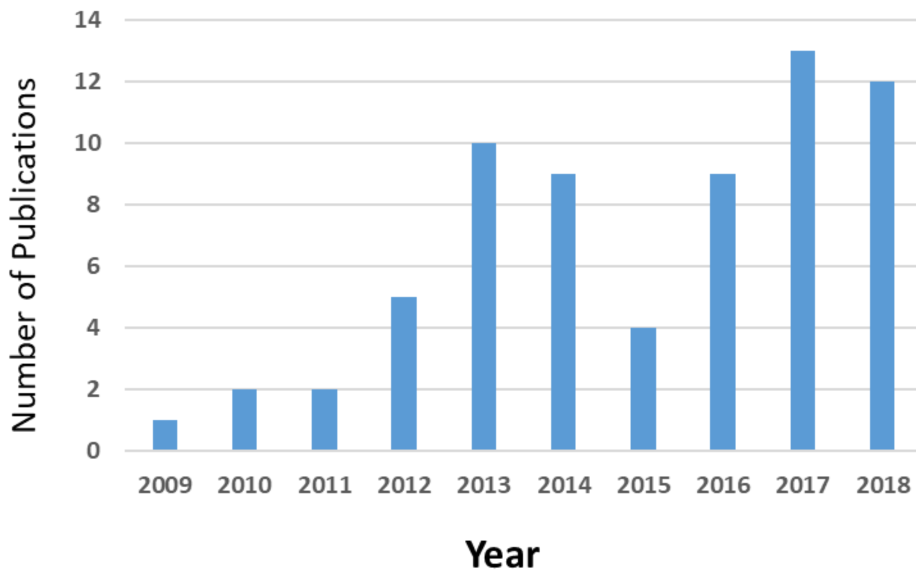

Figure 15: Research trends in electrospinning method in MF technology (total 67 papers)

While majority of the research activity on MF applications is concentrated in wastewater treatment, another application of interest which showed an increasing research trend is the food industry. Figure 16 shows how MF research has evolved over the years in this sector. With only 18 publications in the year 2009, a significant increase to 52 publications was seen in 2018. Apart from some dips in 2012 and 2014, an increasing trend can be observed. This might be attributed to the immediate boost in the food sector, with an instant need for better separation technologies during and after production stages. A recent review by Urošević et al. [59] highlighted the developments in MF of fruit juices and compared it to UF for its advantages, disadvantages and juice clarification. Garcia et al. [60] reviewed MF technology for bacterial removal when applied to dairy streams for improved milk shelf life. The trend in review articles has also seen a positive shift since 2009. This is attributed to the increasing research activity and progress in MF in general. Figure 17 shows that the number of reviews on MF topics was steady with 1 and 4 papers till 2013, after which it saw an increasing trend up to 9 papers in 2016-2018.

A total of 50 review papers were published in the last decade, from which about 44 addressed the issue of fouling in one way or the other [14, 16, 61]. Again, this also signifies the research in fouling in MF membranes which has gained immense importance over the past years.

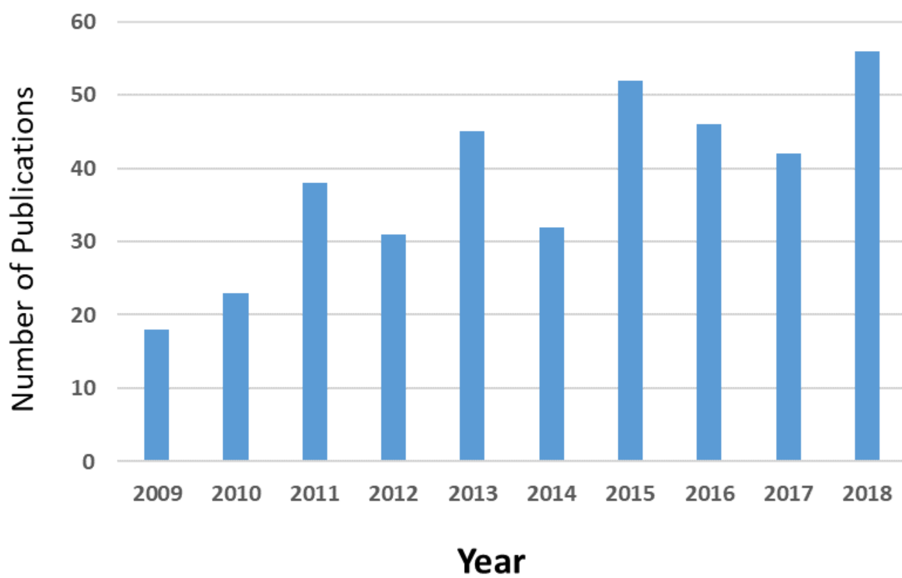

Figure 16: Research trends related to food application in MF technology (total 383 papers)

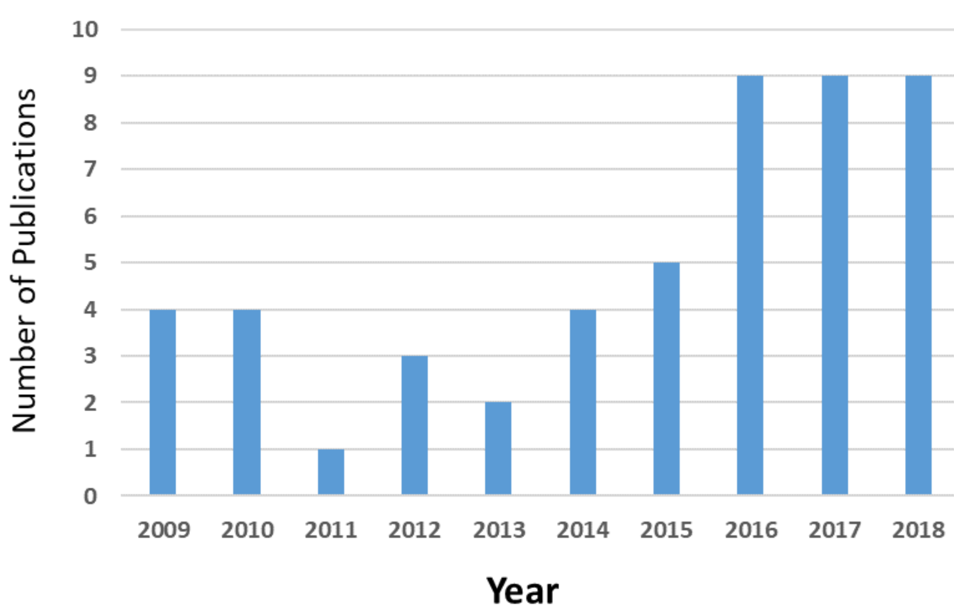

Figure 17: Review articles for MF technology (50 total papers).

5 Decreasing Research Activity

A decreasing research trend was observed in one research area of MF, modeling and simulation. Figure 18 shows the research trend in this area over the past decade. Even though the trend is quite irregular, a general decline can be observed, especially from 2012. The number of publications in 2012 was 21, which declined to 10 and 11 publications in 2017 and 2018 respectively. A definite reason for this decline is difficult to conclude, however, it may be attributed to the limitations imposed by simulating real time conditions during the MF process, leading to complex models and calculations. This is because of the complex feed solutions usually used during MF, such as industrial and pharmaceutical wastewaters composed of several components together. Thus, optimized numerical models are required to successfully model membrane characteristics and MF operating parameters for complex feed solutions. This decreasing research trend is also quite similar to the research trend in NF modeling [62]. Hence, this shows, that this field is somewhat neglected in other membrane-based processes as well, and needs further consideration in integrating experimental and simulation data. Till date, MF modeling includes studying permeate flux behavior [63], particle deposition in the membrane [64], deformation behavior in membranes [65] and identification of optimum operating conditions for simple feed solutions such as green tea extracts [48], beer [66] and bio-ethanol [67].

Classical models usually do not take into account the rate of MF membrane fouling with respect to the flow rate, and thus are inadequate when applied for biological and inorganic feeds. Thus, accurate models are essential for precise modeling. Giglia and Straeffer [68] simulated MF behavior when they were placed in series to each other. The filters fouled at different rates and classical models failed to simulate the effect of flow rate on fouling rate of the membranes. Hence, they extended previous studies on combined intermediate-adsorption fouling models for single to filters to filters operated in series. Their developed model demonstrated improved accuracy in predicting MF performance for bio-fluids, with each membrane performance measured individually.

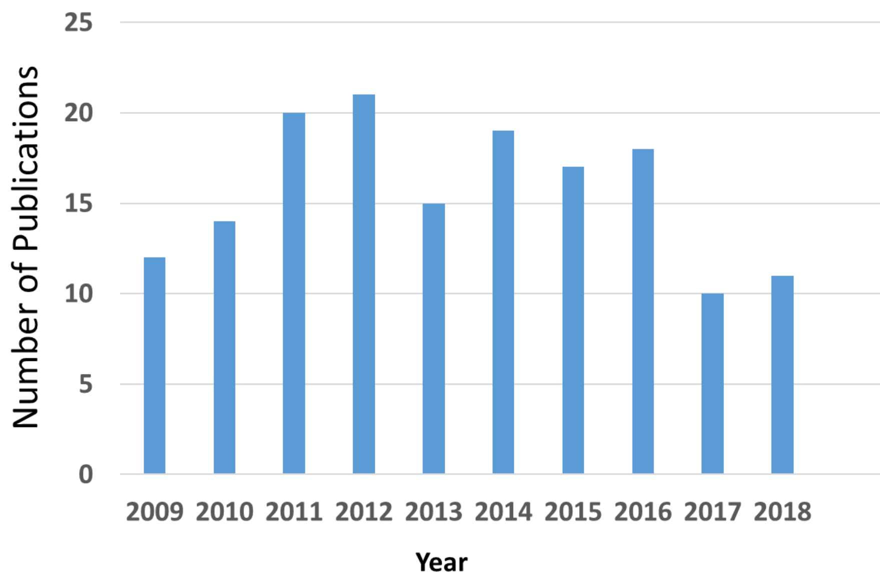

Figure 18: Research trends in modelling of MF processes (157 total papers)

Moreover, as our understanding of modeling builds up, the complexity of handling computational knowledge increases. Several aspects during MF operation may have to be modeled for successful prediction including feed and permeate fluxes, recovery and membrane characteristics. However, majority of research papers have focused on modeling of MF fouling. Rahimi et al. [69] reported computational fluid dynamics (CFD) using Fluent 6.2 code on PVDF membrane using Blue Indigo solution. Their modeling was complemented with experimental investigations explaining shear stress distributions due to the observed membrane fouling. Zare et al. [70] also used CFD modeling for studying concentration polarization in MF of oily water. For the first time, oil concentration was solved using Eulerian multiphase model, which predicted reasonable fouling behavior in different conditions. However, their model required further modifications for a much accurate permeate flux prediction. Liu et al. [71] applied artificial neural network (ANN) models to predict the performance of MF system for water treatment. Membrane performance was evaluated by varying several operating parameters and studying different variables such as permeate flux, turbidity, time and backwash frequency. An accurate agreement between the predicted and experimental values was sought, however, the accuracy of ANN prediction highly relied on the database training. Thus, several limitations like these could be the reason for a decreasing trend in this area of research.

6 Potential Research Areas

While the majority of research activity in MF is concentrated on wastewater treatment, membrane fabrication and modification and on fouling studies, there are some potential research areas which need further research output compared to their existing research trends. One of these is desalination. MF membranes have a large pore size, not sufficient to separate monovalent or divalent cations (see Figure 1). Thus, it cannot be directly employed for salt removal from brackish or seawater. However, MF takes up a significant place in desalination as a pretreatment step, whereby it treats incoming feed water for contaminant removal to improve the feed quality for NF/RO systems. MF has great industrial importance in this sector, whereby many pilot plants are utilizing this technology for improved desalination performance by achieving lower silt density index (SDI). For example, Ebrahim et al. [72] reported a significant improvement in SDI variation of 0.24–3% with an average SDI of 2.42 by using MF pretreatment during their study on Doha Research Plant. Similarly, Corral et al. [73] reported a pilot study on Central Arizona Project water where MF repeatedly achieved a low SDI < 3 which in turn improved the RO plant performance. Figure 19 shows the research trend in the desalination sector employing MF, with 137 papers published in total from 2009 to 2018. There was slight increase from 7 papers in 2009 to 10 and 16 papers in 2010 and 2011. A decrease was seen thereafter till the year 2014. The years, 2016 and 2017 saw a high research output with a total of 49 papers in these two years. Nevertheless, research in desalination again saw a drop in 2018, with only 8 papers published that year. This might be attributed to increasing research activity in NF which has proved its place as a prominent membrane pretreatment to RO, hence replacing MF and UF [62]. Besides, owing to economic feasibility, several conventional pretreatment methods still have a strong hold as a selection criterion for pretreatment. It is worth mentioning that with continuous development in the MF technology, focusing especially on pretreatment of seawater, can lead to big economical savings through biofouling prevention and increased membrane lifetimes. Even with a higher initial investment cost, MF has immense potential to lower down the overall water production cost due to improved permeate qualities. What is required is MF technology with novel backwashing approaches, improved membrane biofouling resistances and increased contaminant selectivity. Due to the variation in seawater qualities across the globe, requirements may vary accordingly. Secondly, development of innovative hybrid processing, such as those combining MF with conventional pretreatments is required.

Thus, research in this direction might be helpful for target applications such as desalination and wastewater treatment.

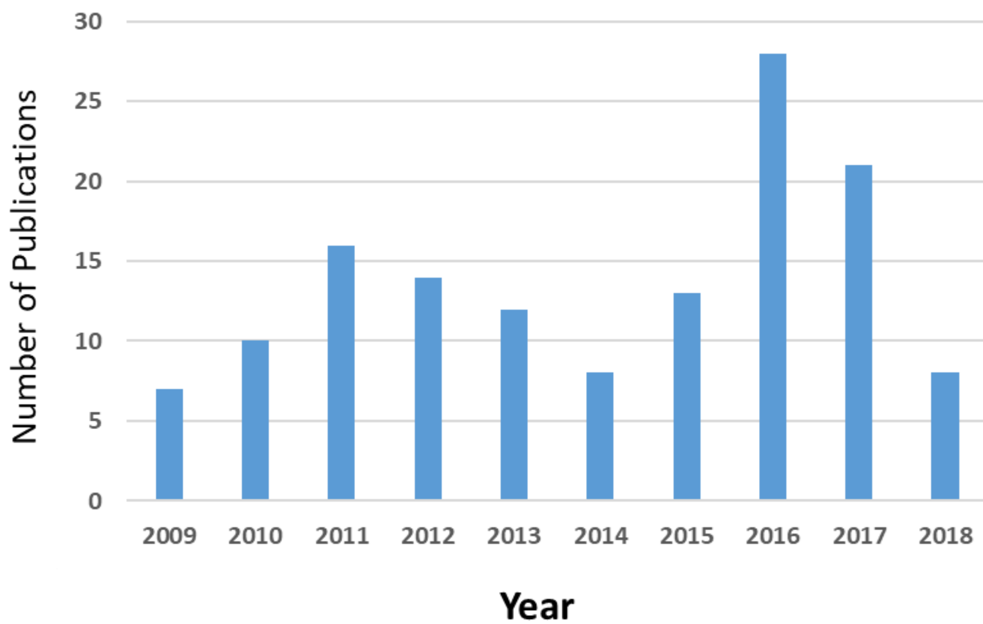

Figure 19: Research trends in desalination using MF (137 total papers).

Moreover, in many cases, MF may be the only solution for selective removal of certain bacteria from dairy products [22]. Otherwise, other processes such as heat treatment might alter the nutritional properties of the milk. However, much research is required in this area where the nutritional value can be truly preserved, at the same time optimizing cost to put a lesser economic burden at the processing stage. This also leads to potential development in novel MF membranes. Though membrane fabrication and modification forms a dominant research area in MF technology, during this review, we found several aspects which were open for further research. Besides the major chemicals identified in Figure 11, there are several others and thus, there is huge potential for numerous membrane materials for optimum MF performances.

Several functional materials may be incorporated in the existing MF membranes for achieving multiple functionalities [74]. For example, the polymeric membrane can be incorporated with various catalytic materials such titania (TiO_2) or zeolite particles for bacterial degradation and improved hydrophilicity. As highlighted in Figure 5, the interdisciplinary nature of MF technology makes it worth considering the progress made in other fields for such catalytic materials when being considered for MF. Such aspects include using nanoscale particles [75], agglomeration problems [76-79], leaching issues [80] and fabrication of hybrid materials for dual functionalities [81-84]. In near future, it will be interesting to see how the progress in membrane development may evolve considering these aspects of membrane fabrication. In addition, fabrication cost is an important factor to be considered. Cost effective approaches need to be developed, both in terms of membrane fabrication, cleaning and overall system. Experimental cost will only be justified if researchers incorporate economics in their studies when they report material development, pilot scale studies and improvements in MF technology.

Another area which needs further attention is the publications on MF reviews. Even though an increasing research trend was seen for review papers (see Figure 17), the progress has remained quite stagnant over the past three years. Moreover, certain areas in MF were identified, which require a complete review. For example, there does not exist any comprehensive review on the fabrication techniques used for MF membranes in specific. Considering the significant progress made this area during the past decade, such a review can be beneficial for researchers who are focusing their efforts for MF membrane development. Other areas for possible reviews include collation of real time pilot studies with different feed water characteristics, and critical analysis on economics and corresponding membrane types and configurations reported.

7 Conclusion

MF research has gained immense importance over the past decade, as evident by the number of publications during the past decade. While the number of publications were constant at 160 papers per year from 2009 to 2011, the numbers increased to about 200 per annum during the past three years. Majority of the published articles were published in The Journal of Membrane Science (22.0%), Desalination (13.0%), Separation and Purification Technology (8.6%) and Water Research (5.3%). Around 51.1 % of the publications are spread in about 150 different journals. Major research topics in MF include wastewater treatment, membrane fabrication and modification and fouling studies. More than 100 papers were published each year on MF membrane development, while about more than 50 papers per annum focused on wastewater treatment and fouling studies. Several research topics were identified with an increasing research trend such as fabrication techniques of phase inversion, coating and electrospinning and MF application in the food industry. in each of these topic areas, publications have more than doubled over the period reviewed. Moreover, there was an increasing trend in the number of review papers from 2009 to 2015, after which the numbers became stagnant at 9 reviews per annum. Thus, there lies a much greater potential for researchers to collate certain research studies in MF for reviews, to benefit the various industries utilizing this technology. Besides increasing trends, a decreasing research trend was identified in the area of modeling and simulation, which might be attributed to the complexity of numerical models and failure of classical models for difficult feed waters encouraging membrane fouling. Nevertheless, being an important membrane separation technology, interest in MF continues to grow with several potential research areas yet to be fully explored.

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