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# A Review of Efforts To Reduce Membrane Fouling by Control of Feed Spacer Characteristics

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## Highlights

- Aspects of feed spacer modification are examined in this comprehensive review
- Appropriate feed spacer design can do much to mitigate fouling in membrane modules
- Continued effort is needed towards the enhancement of high flux and low-pressure drop in membrane modules
- More work is needed on the relationship between feed spacer modification and underlying membrane performance

## Abstract

A major problem for the operation of membrane based water treatment systems is fouling of the membrane surface. Within industrial modules, such as spiral wound modules, feed spacers are necessary to separate adjacent membrane layers. Engineering of feed spacers with novel characteristics is an attractive option for reducing fouling within the module, for instance by increasing surface shear forces. This paper reviews the characteristics of feed spacers which are important in fouling and efforts to design novel feed spacers with anti-fouling characteristics, such as by addition of surface coatings, modified feed spacer design and geometric considerations, 3D printing and use of electrically conductive feed spacers. This paper also provides a brief review of techniques available to assess aspects of feed

spacer design. It is seen that appropriate feed spacer design can do much to mitigate fouling in membrane modules, but continued effort is needed toward enhancement of high flux and low-pressure drop in membrane modules.

Keywords: Feed spacer modification, feed spacer characterisation, membrane module, fouling mitigation, flux, spiral wound module, membrane fouling.

## **1.Introduction**

Successful application of membrane technology necessitates not only high membrane performance, but also the engineering of feed spacers which can effectively manage fluid-flow at the membrane surface. A major contributor to flux decline during membrane separation processes is concentration polarization (CP) of the solute, in conjunction with irreversible fouling at the membrane surface [1,2]. Both cross-flow profiles and permeate flux are altered by concentration polarization [3,4]. Fluid-flow management determines optimal membrane module performance, since it effectively enhances fluid-flow [5-7]. From an operational perspective permeate flux, cross-flow, module geometry and array shape control the extent of CP [8,9]. In addition CP has a major effect on inorganic fouling of the membrane surface, due to accumulation of solute happening at a greater rate than back diffusion to the bulk solution. To overcome these problems, a number of research strategies have focused on adapting feed channel spacer in membrane models to enhance membrane performance [10-14].

Feed channel spacers are an essential part of spiral wound module (SWM) design. The feed spacer ensures inter-membrane spacing and improving mixing [15,16], and has a major impact on membrane performance [7,17]. However, the feed channel spacer may offer localized dead spots with poor mass transfer that encourage fouling[18,19]. Thus, for better feed spacer performance, the designers should attempt to maximize the mass transfer whilst at the same time minimizing the pressure drop, as this will reduce fouling initiation [2]. However, the most significant impairment to the efficient operation of membrane units is membrane fouling, which seriously hampers the application and uptake of membrane technologies [20,21].

Fouling is a complicated phenomenon determined by the interplay of several mechanisms whose activity varies depending upon the specific conditions [22,23]. Fouling is defined as the detrimental deposition of retained particles, colloids, salts and macromolecules at the surface (surface fouling) and/or inside the membrane pores (internal fouling) [24-28]. Generally, several categories of foulants and fouling can be defined, depending on the physical and chemical nature of the foulant species: inorganic (scaling), organic, colloidal and biofouling [29-32]. The accumulation of foulants can come in the form of concentration polarization, physical pore blocking or cake/gel layer formation [56]. Membrane fouling sets-up a barrier layer or film on the surface of the membrane or blocks the membrane pores leading to inhibition of water flux, increasing the pressure drop across the membrane and reducing permeate productivity [33-35]. In spiral-wound membrane modules, pressure drop can be classified into two categories: the feed channel pressure drop (the pressure drop between feed and brine lines) and the transmembrane pressure drop (the differential pressure between feed and permeate lines). With the present of a feed spacer, during biofilm accumulation and biofouling generation, an increment of transmembrane pressure drop

occurs and the membrane flux declines. The impact of membrane fouling on the development of transmembrane pressure drop is insignificant in comparison with the transmembrane pressure drop due to feed spacer fouling [19,68]. Much research into fouling of membrane modules focusses on biofouling of the modules as a whole, rather than looking at specific interactions with the feed spacers directly, or at other types of fouling [118]. The consequences of fouling and resulting decline in flux ratio leads to an increase in the operating pressure necessary to maintain permeate output, as well as requirements for extensive pre-treatment and cleaning procedures and chemicals, leading to an increase in operating and maintenance costs of the affected water treatment system [36,25]. For these reasons, membrane fouling is a significant concern when considering the engineering aspects of membrane technology [37,38].

Organic fouling progression occurs due to adsorption is considered the most commonly mechanism conjoined with organic fouling and therefore, interaction between the organic foulant and membrane surface is of supreme significance issue. Biofouling is considered more specially challenging due to the prospect for membrane pores to become blocked. Also, biofouling can assistance the other types of fouling, such as inorganic fouling, as these channelling matters leads to the precipitation of soluble salts and, eventually, scaling [57].

During industrial scale membrane processes, the exclusion of contaminants in order to gain clean water is most often performed by means of SWM modules. Accordingly, optimisation of the performance of these membrane modules has been focussed on the development of membranes [39] and engineering of the feed channel spacer design [40]. In the case of feed spacer optimization, the reported research works can be classified into two categories: (a) efforts to enhance the conventional plastic spacer design by studying the impact of the spacers orientation and geometry, for instance spacer thickness, filament diameter, and mesh length on water treatment process performance; (b) novel feed spacer designs which can surpass the conventional feed spacer performance [41]. The majority of feed spacer modification investigations are either experimental or numerical modelling studies aimed at estimation of the underlying phenomena and with optimizing the feed spacer configuration [42]. The effect of hydraulics on spiral-wound membrane systems performance as well as electrically conductive spacer meshes, for application of effective cleaning strategies using electrolysis have been of much recent interest [43,44]. Approaches for controlling biofouling focussing attention on the feed spacer have comprised changing the feed spacer thickness or orientation [45], periodic air/water cleaning [46], and coating of biocides on the net-spacer [47,48].

Consequently, it is essential to provide an up-to-date review of feed spacer modification and its role in improving membrane fouling control. There are a large number of studies that have adopted different approaches in the development of novel membrane modules for water treatment applications. However, the focus of this review is concentrated on the modification of feed spacers. This includes firstly looking at the fundamentals of feed spacer configuration, followed by other important aspects such as feed spacer modification and applications for membrane fouling mitigation. Finally, recently reported techniques to characterize feed spacer surface morphology and its relationship to fouling are summarised, with a discussion of the main issues and challenges which need to be addressed in future investigations (see: Figure1).

## **2. Effect of feed spacer modification on fouling**

The conventional commercial membrane configuration is the SWM, which presents a large membrane surface area. This configuration includes polymeric separation grids, which act as

spacers between adjacent membrane sheets, keeping an open flow path while also offering flow disturbance that may contribute to decreased CP by improving feed mixing. Alteration of feed spacer geometry is a potential option to reduce the impact of fouling on the performance of membrane systems. Commercially, there are two main feed spacer configurations available: woven and nonwoven [50,51]. Good spacer configurations should reduce a build-up of fouling deposits and decrease CP via keeping the solute concentration in the layer of fluid in contact with membrane surface at close to the bulk concentration [52,53]. Rejected species accumulation can be suppressed by encouraging back-mixing from the solution layer adjacent to membrane to the liquid bulk [50]. An emerging methodology is to modify feed spacer configuration to reduce fouling. Generally, several strategies have been adopted to develop mesh-type spacers such as by surface coating[54], altered geometry design [55] or three-dimensionally printed feed spacers [41], and use of electrically conductive spacers[43].

## **2-1 Effect feed spacer surface coating modification.**

Many approaches have been adopted to modify the features of feed spacer surfaces through coating as a strategy for improving filtration performance and reducing fouling impact. Several attempts have been reported in this field, for example Hausman *et al.* [56,57] studied the efficiency of the biocidal properties of polypropylene as a membrane feed spacer for reverse osmosis, wherein a spacer arm was functionalized with chelating ligands charged with the ions of copper to purify water from microbial biofilms. Modified and Virgin films were submerged in solutions in contact with  $3.0 \times 10^5$  *Escherichia coli* /mL . Fouling analysis revealed that the number of cells adhered to virgin sheets after 7 days was an order of magnitude greater than those attached to the modified sheets.

Yang *et al.* [48] investigated the potential of biofouling control through the membrane and feed spacer surface modification with a nano-silver coating. Silver nanoparticles were directly coated on the reverse osmosis membrane and feed spacer surface using a chemical reduction technique. The antifouling performance of modified membrane with unmodified feed spacer as well as the coated feed spacers along with a virgin membrane were tested in a cross-flow cell, and their fouling control performance compared. Permeate flux decline, salt rejection and microbial activity progress on the membrane cell were monitored and quantified. They stated that the analysis results exhibited that both coated membrane with uncoated feed spacer and coated feed spacer with unmodified membrane presented better than the uncoated membrane and feed spacer in terms of decline in permeate flux and salt rejection. Also, they stated that the effect of coated feed spacer on antimicrobial activity was more durable (see. Figure 2).

Araújo *et al.* [47] investigated the effects of metal coating on feed spacers in commercially spiral wound membrane modules and studied their potential for anti-biofouling activity. The influence of metal coating on biofilm activity was examined with six parallel-operated membrane fouling simulators. Copper and silver were adopted as biocides for the control of biofouling. The effect of gold coating was also employed to compare with silver and copper coating. Surprisingly, they noticed that the feed spacers coated with copper and silver did not reduce the fouling rate compared to the uncoated. The lowest feed channel pressure rise was observed for the gold coated feed spacer. Biofilm characteristics were measured by adenosine tri phosphate (ATP) (see Figure 3-C) and total organic carbon (TOC) levels (Figure 3-D and Figure 3-C). Based on observations, the authors concluded that the impact of feed spacer modification with toxic compounds such as copper and silver had a slight effect, but were not sufficiently effective to prevent biofilm formation. The best performance was gained with gold coated feed spacer. However it was concluded that the antibiofouling effects were

insufficient to provide an economical way to reduce the impact of biofouling on membrane performance.

The potential of polydopamine-g-poly(ethylene glycol) and polydopamine coating for biofouling control with polysulfone ultrafiltration, TS80 nanofiltration membranes and feed spacers was investigated by Miller *et al.* [58]. The fouling propensity of coated and uncoated membranes and spacers was assessed experimentally over several days in membrane fouling simulators, wherein *Pseudomonas aeruginosa* and bovine serum albumin were employed as a typical biofouling. The analysis of experiments was demonstrated that polydopamine-g-poly(ethylene glycol) and polydopamine-coated membranes and feed spacers showed significantly reduced adhesion of *P. aeruginosa* and bovine serum albumin in the short-term during adhesion tests. Meanwhile no reduction of biofouling was observed during long-term experiments with modified ultrafiltration, nanofiltration membranes and feed spacers. These results revealed that polydopamine-g-poly(ethylene glycol) and polydopamine coatings are ineffective at preventing biofouling.

The anti-biofouling performance of feed spacers coated with zinc oxide nanoparticles was studied by Ronen *et al* [59]. Modification of the feed spacer was achieved via a sono-chemical deposition technique. The experiments were carried out using a polysulfone ultrafiltration membrane combined with the modified feed spacer under flow conditions in a membrane fouling simulators system, wherein *Pseudomonas putida S-12* was used as a model biofoulant. An antibacterial nano-composite coated spacer was evaluated and examined for control of biofilm formation and the results of the experiments exhibited a reduction in attached bacteria to the surface of the modified membrane-feed spacer. AFM analysis of the modified polypropylene spacer revealed an electrostatic repulsion which could also reduce bacterial adhesion.

A similar effect was investigated by the same group when using nano-silver coating to modify feed spacers [54]. Numerical simulation was used to estimate the distribution of silver-ions in the feed channel and close to the ultrafiltration membrane surface. The nano-silver was inserted into the polypropylene mesh using a sono-chemical deposition method. Biofilm accumulation using modified feed spacers was monitored experimentally using a crossflow filtration set-up, and compared with attached biofilms at unmodified feed spacers. The results showed that the membrane surface underneath the unmodified feed spacer exhibited large clusters of bacteria, and only 12% of bacteria were dead. Conversely, the membrane surface underneath the modified feed spacer presented considerably less bacteria attached to the membrane surface: 27% were of bacteria was dead (see Figure 4). Hence, the silver coated feed spacers demonstrated enhanced and steadier permeate flux during ten days of experimental operation compared to the control one.

However, the antifouling effects on the surface modification of feed spacers and or biofilms have been found by several researchers to not be sustained over a long period of time due to covering of surfaces by conditioning films or degradation of the surface modification [60][45][55][61].

## **2-2 Effect of geometrical modifications of feed spacers**

Research into feed spacer design has mainly focused on the impact of spacer geometry on fluid dynamics and mass transfer in the membrane system. Geometric modification of feed spacers includes altering the spacer thickness, the internal strand angle, and spacer mesh size (the distance between the strands) [62]. In addition to this, the feed spacer strand shape can be modified [55]. The major aim of geometric modification of the feed spacers is to reduce the effect of fouling on the membrane system without compromising water production [63].

Research into the alteration of feed spacer geometry has been carried out, either experimentally or using computational modelling to gain insights into the impact of hydraulics on the spiral-wound membrane systems [64] and the potential of design changes to decrease the effect of fouling, particularly biofouling on membrane performance [55], also the potential to develop novel effective self-cleaning strategies[54]. Several surveys have monitored flow characteristics to compare effects of different geometries of feed spacers. In essence the presence of feed spacers allow secondary turbulent flows to allow improved mixing of fluid adjacent to the membrane surface back to bulk solution, preventing build-up of solute and foulant concentrations [111,114]. Essentially, efforts to modify geometry of spacers to reduce fouling by improving these mixing mechanisms. This section deals with research into using novel geometries to reduce module fouling.

### **2-2-1 Modification of feed spacer design**

The investigation of feed spacer design has shed light mainly on the impact of the feed spacer geometry on feed channel hydrodynamics, which in turn significantly affects all other parameters. The feed spacer geometry determines the power necessary to overcome the fouling-derived resistance to fluid-flow. The dissipation of the yield flow pattern is associated with the shear stress distribution on the feed spacer filaments and membranes [5], which has effects on the permeate flux [65,66], scaling [67], biofouling and operation [68]. Thus, these phenomena should all be considered in any proposed new designs intended to optimise feed spacer geometry to reduce fouling[69].

Numerical modelling is a powerful tool to evaluate the impact of design parameters on filtration process performance, for a varied range of operating conditions carried out in SWMs. Computational fluid dynamics applied to solute transport has been used in various investigations to model flow and permeation, attempting to acquire the optimal design of feed spacer [70,71]. Ranade and Kumar [72] found that when modelling unit cells in SWMs, flow fields were little different between curved or flat spaces when the spacer is present. This is because secondary flows caused by curvature are insignificant compared with those generated by the spacer geometry. These results were in accordance with previous experimental observations by Schock and Miquel [69]. It was concluded by the authors that this indicated that mass transfer and pressure drop data derived from experiments with flat membrane surfaces were suitable for describing the situation in curved SWMs.

In addition to three-dimensional models, computational modelling has been applied to two-dimensional investigations in influence of alignment and shape of spacer strands. The majority of the modelling in two-dimensions have been performed using a ladder spacer strand pattern, where the feed spacer strands are aligned transversally and axially to the bulk flow direction. The impacts of strand size and alignment on scaling and (bio)fouling in reverse osmosis for desalination have been subject of study [67]. Even though cylindrical strands have most commonly been taken into inconsideration, other cross-sectional forms, for instance triangular, saw-tooth [73], square [74], and elliptical [75] have also been assessed.

Simulations in two-dimensions have accelerated the computational and qualitative examination of the design parameters for ladder-type feed spacers[66]. However, it cannot be utilized for quantitative appraisals of spacer designs, in which the strand geometries are symmetric in the direction lateral to the bulk flow in two-dimensions. With greater computational resources, the analysis of various feed spacer designs in three-dimensional models has been able to progress without this limitation. Evaluation in three-dimensions has been done for several filtration processes, including reverse osmosis and ultrafiltration [70].

Three-dimensional modelling has led to the design of novel feed spacers with strand cross-sections that are other than circular [72,76].

The majority of investigations into optimal feed spacer design have focussed on the effect of alignment of cylindrical feed spacer filaments: flow attack angle,  $\alpha$ , and internal strand angle,  $\beta$  and the influence of their dimensions with respect to their mutual distance as well as channel height on both hydrodynamics and solute transport [72,77,9]. Modelling of biofouling on feed-spacer filaments found this to have a greater effect on the assessed pressure drop than fouling on the membrane itself [57,65], that led to a shift in focus to the importance of optimal feed spacer design.

Gu *et al* [78] studied four different categories of feed spacer configuration with a total of 20 geometric differences (see Figure 5) The impact of feed spacer design on membrane performance was examined using three-dimensional computational fluid dynamics simulations. Considering the adopted operating condition and the net spacer geometries it was concluded that fully woven spacer geometries outperform counterpart spacer configurations, in terms of higher water flux, and lessening the concentration polarization. Even though the related pressure drops,  $\Delta P$ , are slightly more than their nonwoven equivalents, reducing spacer net angle leads to decreased water flux and extended concentration polarization magnitude. In addition, the  $\Delta P$  is extremely sensitive to the attack angle (the angle between axial filaments and the direction of inflow).

Kim *et al* [79] investigated two spiral wound forward osmosis modules containing cellulose tri-acetate and thin film composite membranes, in terms of hydrodynamics, fouling behaviour and cleaning strategy. For both modules, a 5-times lower flow rate was demanded in the draw than needed in the feed channel, attributable to significant pressure drop in the draw channel, and was a remarkably critical operating challenge in the cellulose tri-acetate module when permeate spacers were employed. They concluded that the feed spacer design plays an important role in pressure drop in the spiral wound forward osmosis module. In that aspect, the diamond feed spacer used in the thin film composite module was observed to be less restrictive to flow than for the cellulose tri-acetate module (the spacer had a thick tricot-type weave which created more resistance to the flux). However, less mechanical support was offered to the feed stream in the module (See Table 1).

Taamneh Y. and Bataineh K. [80] improved the performance of direct contact membrane distillation by utilizing a spacer-filled channel. The impact of the spacer filament's orientation on the flow pattern in a direct contact membrane distillation module was examined numerically using computational fluid dynamic simulations and by experiment. Spacers were used where the top filaments were oriented at an angle of  $30^\circ$ ,  $45^\circ$ ,  $62^\circ$  and  $90^\circ$  respectively relative to flow, with the bottom set parallel to the flow channel axis. In addition, a spacer was used where both sets of filaments were oriented at  $45^\circ$  to the channel direction (see Figure 6). It was found that the spacer filament's orientation effected the heat transfer and fluid-flow performance significantly. The feed spacer filaments oriented at an angle of  $45^\circ$  to the channel axis yielded the highest predicted average shear stress and showed the best performance in reducing fouling accumulation.

Kavianipour *et al* [52] used computational fluid dynamics to study four feed spacer configurations: triple, ladder-type, wavy and submerged long with spiral wound modules that are employed in reverse osmosis systems (see Figure 7). Both mass transfer phenomena and flow characteristics were investigated and the feed spacer behaviour was compared at different Reynolds numbers in the range  $50 \leq Re \leq 200$  through the laminar zone. The spacer configuration results for the four feed spacer geometries revealed the ladder-type was the best



choice for Re below 120, whereas the wavy-type showed the best results for Re greater than 120. Thus, the adopted wavy-type with increasing Re value is more beneficial.

Sidiqi *et al.*[55] investigated interplay between the hydraulic resistance of clean feed spacers and the potential of biofouling on feed spacers under controlled conditions involving investigation of biofouling during short (9 days with nutrient dosing to accelerator biofouling) and long-term (96 with no added doses of nutrient) membrane system operation, as shown in Figure 8. They evaluated the potential of biofouling for two commercially available feed spacers (the same filament shape and internal strand angle (90°) and spacer thicknesses of 0.86 and 0.79 mm and four modified feed spacers (modified inner strand angle and thinner strands compared with commercial spacers; modified larger mesh-size and thinner strands; modified alternate strand thickness with irregularities in strands; modified alternate strand thickness). The pressure drop of the six feed spacers were compared at different flow rates. It was found that the modified feed spacer designs exhibited a lower pressure drop for the same flow rates compared to the commercial feed spacers. However, the trend observed for hydraulic resistance was markedly different to the trend seen for biofouling resistance. The impact of biofouling was identical for short-term biofouling studies (with nutrient) as for long-term biofouling investigations (without nutrient).

When optimising membrane modules to handle poor quality feedwater, which tends to cause clogging in the spacer, thicker spacers have been found to reduce pressure drop due to fouling. To counteract the negative effects of spacer thickness on available membrane area, new strategies for gluing membrane leaflets have been adopted by manufacturers [81]. Porosity and spacer geometry impact on the fluid-dynamic behaviour in feed spacer-filled channels [82]. To identify the reason for pressure drop reduction the spacer-filled channels' geometry and porosity should be determined for each of the feed spacers employed.

### **2-2-2 Three-dimensional printing of novel feed spacer designs**

Many prior investigations have concerned themselves with optimizing the geometry and orientation of feed spacers. There is an essential requirement for a feasible technology to fabricate these innovative spacer designs for practical testing and validation. The three-dimensional printing technique, known as additive manufacturing, allows the relatively easy manufacture of forms which are inaccessible using conventional fabrication methods [49]. It allows the engineering of freeform objects layer by layer using a variety of different materials, such as metals, polymers, composites and even novel materials [41]. Industrially, there are several applications for objects fabricated using the three-dimension printing technology: for example, marine and offshore [87], building and construction [88], biomedical [89], food industry [90], as well as desalination and water treatment [41]. The main advantage of three-dimensional printing is that it allows the easy engineering of patterns with the complicated geometry [49]. Figure 9 gives a historical summary of milestones of three-dimensional printing membrane module design.

This technology now allows investigators to freely produce novel spacers with complex geometries that were previously limited by traditional manufacturing procedures. The function of the feed spacer in SWMs is enhanced mass transfer and reduction of the impact of concentration polarization. However, the interaction between the feed spacer and the membrane can lead to increased fouling of the membrane because of the spacer shadow effect, where the movement of solute ions is reduced in close proximity to the spacer [68]. By a combination of three-dimensional printing techniques with the optimal design of membrane modules, the membrane fouling issue could be addressed via the optimization of feed spacers to increase mass transfer and decrease the polarization. However, the three-dimensional

printing method is not perfect, for not all design features can be additively manufactured with accuracy and this lack of accuracy may inadvertently result in various derivations in the surfaces and geometry from what is intended. Development of three-dimensional printing tools in terms of materials, resolution, and speed should ensure the production of many highly efficiency membrane module components [49].

The first three-dimension printed feed spacers were described by Li *et al.* who used a powder based Selective Laser Sintering process [91]. Their feed spacer designs included three different feed spacer modifications: modified filament feed spacer, a helical feed spacer and multi-layered feed spacers. The various fluid-flow patterns generated by the different designs and their impact on mass transfer were addressed. They stated that obtaining accurate mass transfer simulations from computational fluid dynamic was complicated due to the feed spacer engineering designs. Balster *et al.* also, studied the potential of multi-layered spacer structures for enhancing mass transfer in electro-dialysis systems[92], by analytically investigating the influence of various geometrical parameters of single feed spacers, comprising different filament sorts (helical vs. normal), mesh lengths, filament diameters and fluid-flow attack angles on the mass transfer. These feed spacers, shaped by Selective Laser Sintering, were used to study concentration polarisation in electro-dialysis desalination processes. Experimentally, the feed spacer performance was determined via limiting current density at a range of flow velocities to allow measurements of mass transfer effects and cross membrane power consumption. For single layer spacers, those with twisted filaments and attack and filament angles of  $60^\circ$  showed the highest mass transfer, which was attributed to their promotion of swirling flows. However, multi-layered spacers show the best performance. The multi-layer structured feed spacer with smaller middle layer filament diameter was able to yield a mass transfer that was 20% higher than seen with common mesh spacers.

Three-dimensional non-net type feed spacers were first described in 2008 by Shrivastava *et al.*[93] who developed them for application in ultrafiltration and reverse osmosis systems, using a solid-based 3D printing method: the Fused Deposition technique. A number of designs were examined, including helical feed spacers as well as asymmetric structures such as laddered and herringbone type feed spacers in order to study the impact of feed spacer shape in decreasing concentration polarisation and to provide an optimised feed spacer design. Mass transfer rates were measured using electrochemical techniques and demonstrated enhanced flow which was least for ladder shaped spacers, was intermediate for asymmetric herringbone shaped spacers and the most for helical spacers. However, the authors highlighted some concerns with their methodology, which centred on the electrochemical techniques being diffusion based, which may not correctly apply to the flow based pressure driven filtration processes, for measurement of mass transfer effects, although as concentration polarisation is largely a diffusive process the issue would not be so damaging.

A feed spacer for spiral wound module inspired by static mixers was invented by Liu *et al* with a liquid based three-dimensional printer known as the Stereo-Lithography Apparatus [94]. The feed spacer was engineered to allow fluid in the upper and lower parts of the fluid flow channel to replace and be replaced by the fluid flowing in the centre of the channel, acting like a static mixer for planar flow [49]. A theoretical model was developed and employed to predict the impact of the adopted feed spacer on both mass transfer and pressure drop which then was validated by ultrafiltration experiments. Despite the pressure drop being higher than that of the typical conventional feed spacers, under similar conditions, the static mixing feed spacers revealed comparable mass transfer at high flow-rate and better mass transfer at low flow-rate. After the micro-structured dual-layered feed spacers was fabricated

by Liu *et al.*, Fritzmann *et al* used the Polyjet printing method for air-sparged-submerged membrane modules [63]. Fritzmann *et al.* [95] continued with examination of these feed spacers in terms of their effects on mass transfer and separation features: for instance, selectivity in ultrafiltration processes. The authors showed that the spacer geometries significantly impact the selectivity, which could have a great effect in fractionation processes using ultrafiltration membranes. Considerable enhancement in both mass transfer and selectivity of the feed spacers compared with that of conventional feed spacers was also reported. In parallel, there was an investigation to compare the impact of additive manufacturing techniques, Fused Deposition Modelling and Polyjet in the manufacture of feed spacers by assessing the morphology and dimensional accuracies of the printed feed spacers [49]. However, the feed spacer functionality was not evaluated.

Even though there are a wide range of commercial three-dimensional printing techniques, only Selective Laser Sintering, Fused Deposition Modelling, Stereo-Lithography Apparatus or Polyjet have been used to fabricate feed spacers. This is owing to the main advantages of these methods: non-support removal and non-toxicity for Selective Laser Sintering, soluble support for Fused Deposition Modelling and Polyjet and good resolution of Stereo-Lithography Apparatus size. Siddiqui *et al.*[64] proposed an approach for development, characterisation, and evaluation of the feed spacers computationally and experimentally. All the adopted feed spacers had an identical geometry with strands differing in their thickness ranging between 0.66 to 0.86 mm (see Figure 10). Based on the numerical modelling results, the analysis showed that the impact of three admission printed feed spacers on hydrodynamics and biofouling can be enhanced. A tuneable relationship between the measured linear flow velocity and pressure drop for feed spacers was observed with similar geometry, demonstrating that spacer modelling can perform as a primary step in spacer characterization. The authors declared that three-dimensional printing technology is an appropriate tool to fabricate feed spacers with a thin and complex geometry: all the adopted 3D printed spacers with identical geometry exhibited similar hydrodynamics and biofouling development. In comparison with the reference feed spacer, a three-dimension modified-geometry printed spacer showed a lower pressure drop as well as lower biofouling influence on spacer and membrane performance.

Tan *et al* [41] investigated several three-dimensional printing methods that result in different spacer geometry and surface morphology and their impact on membrane performance and fouling-mitigation. It was stated that all the printed spacers by powder-based Selective Laser Sintering, solid-based Fused Deposition Modelling, and Polyjet showed improved mass transfer at normal and critical flux than the commercial feed spacer (see Figure 11). However, the microstructures may have a critical effect on the hydrodynamic conditions in the micro-environment and thus affect the concentration polarization. In addition, the bacteria attachment affinity can also be affected by the surface finish. The implication of this study is that parameters to be considered in spacer fabrication by three-dimensional printing method, such as accuracy, geometry, and surface finish associated with the construction techniques are equally important.

Several essential challenges need to be investigated before 3D printing technology can be applied successfully for the engineering of membrane modules. In addition to resolution, a limited range of materials have been used for the 3D printing of feed spacers, and high material costs might be one extremely challenging drawback. Consequently, consideration for alternative approaches to overcome these obstacles is required to attain the optimum potential of 3D printing technology for feed spacer design and fabrication.

## **2-3 Electrically Conductive Feed Spacers**

Among the approaches to improving membrane anti-fouling performance, much focus has been shifted towards using electrochemical systems in conjunction with water treatment technology, which allow for *in-situ* cleaning of membrane systems using electrolysis. For this purpose, an electrochemical cell is used, which comprises anode and cathode electrodes, in which the separating feed solution performs as an electrolyte. However, *in-situ* membrane control of fouling, whilst showing great promise, is very challenging [83-85]. Accordingly, several studies have highlighted the use of an electric current to prevent aquatic fouling on an electrically conductive nanocomposite membrane. Noticeable flux recovery was observed, which was attributed to the electrostatic repulsion between the foulant and the modified membrane surface. Unfortunately, it has been found that the durability of thin active layers at the membrane surface is inadequate due to erosion by water flow. With membranes fabricated using the phase inversion method it is difficult to have optimum morphology due to the additives which are imparted to enhance membrane conductivity. In terms of engineering of membranes using the electrospinning method, the major obstacle is the difficulty in gaining a suitably small pore size. As a result, application of the electrical potential to the electro-conductive feed spacer, rather than the membrane surface, is a promising alternative modification, wherein foulant mitigation either through oxidation (spacer acts as an anode in electrochemical system) [84] or bubble generation (spacers acts as a cathode) [85,86] to cause disruption and release of fouling layers. However, self-cleaning membrane fouling mitigation very challenging [43]. Baek *et al* [44] used a lab-scale cross-flow setup with an electro-conductive feed spacer to simulate a SWM employed in industrial applications. This comprised a two-electrode system consisting of a titanium mesh and a permeate SUS mesh. A suspension of the bacterium *Pseudomonas aeruginosa PA01 GFP* (initial concentration  $1 \times 10^7$  CFU/mL) was used as a model biofouling organism. Membrane biofouling was achieved in the following steps: firstly the membrane was allowed to compact by filtration of deionized water for 18 hours; then conditioning by the filtration of feed solution (10mM NaCl, 10 mM Na citrate ,0.1% soy broth) for about 6 hours; after that biofouling with the *P.aeruginosa PA01 GFP* was applied for 24 hours. After biofouling (biofilm layers growth) had occurred, the electro-conductive feed spacer was polarized via application of either positive, negative or alternating potential for half-hour. The permeate flux recovery was evaluated with the feed solution for a further 3 hours. As shown in Figure 12 after biofouling the permeate flux was reduced to about 47% of the non-biofouled flux. When electrochemical treatment was applied, the flux was recovered to a significant extent.

Abid et al [43] investigated the ability of the application of electrical potential to a coated plastic feed spacer to function as a tool for controlling organic fouling in a lab-scale cross-flow membrane system. The plastic feed spacer was made conductive through dip coating with a carbon-based ink containing graphene nanoplates (GNPs). In the electricidal system, the conductive spacer acted as a cathode and a graphite electrode performed as a counter-electrode (anode), meanwhile the suspension of sodium alginate performed as the aqueous electrolyte solution. When an electrical potential was applied, micro-bubbles were generated at the spacer net, allowing *in-situ* membrane fouling mitigation with noticeable flux enhancement through repeated cycles without any damage to the membrane surface. A significant comparison was made for the electrochemical and *in-situ* cleaning behaviour between titanium metal and coated spacer. Sodium alginate suspension of 20 ppm was used as model organic foulant, which was evaluated at an operating pressure of 0.5 bar as a function of time. Figure 13 shows the comparison between relative flux for both titanium spacer and coated spacer at 30, 45 and 60 min filtration intervals. The results suggest that an electrically conductive feed spacer with an appropriate electrical potential is an efficient approach for organic fouling control in membrane systems for water treatment. Accordingly, there is no need use for chemical reagents or membrane backwash, which traditionally

shortens the membrane modules life span. Additionally, electrically conductive spacers can be easily integrated with all types of pressure driven membrane comprising: MF, UF, NF and RO modules.

It has been reported that periodic electrolysis is considered as an efficient and fast *in-situ* cleaning technique for conductive substrates via bubble generation and has the potential to revolutionize the current performance of membrane modules and could decrease energy consumption and chemical reagent usage in water treatment plant operations [55,83,86].

### **3. Techniques for feed spacer functional characterisation.**

Membrane module efficiency is restricted by concentration polarization and fouling-generation, so the development of methods to mitigate these negative impacts are required through investigation of the fluid dynamics through the feed spacer interstices. A better understanding of the underlying mechanisms of feed spacer performance are necessary to bridge the gap between theoretical predictions and experiment. Researchers have adopted several methods to characterise fluid-flow within feed spacer channels in membrane systems.

#### **3.1 Imaging Techniques**

The particle image velocimetry method was utilized by Gimmelshtein *et al.* to examine the fluid velocity distribution through a membrane system [96]. The particle was modified to investigate two-phase fluid dynamics by Willems *et al.* [97]. The direct observation technique through the membrane module was also harnessed to show the interplay between the feed fluid and the net-spacer: the movement of tracer particles and microorganisms [98,38], micro-bubbles [99], and latex beads [100,101] inside the feed spacer have been analysed. However, such visualization methods are unable to resolve the variation of the fluid-velocity field in the direction perpendicular to the membrane surface, which is needed to clarify the hydrodynamic behaviour adjacent to the membrane.

Alternative imaging methods, such as scanning electron microscope, can offer a high image resolution. However, it is a destructive technique, the visual area is small and comprises only a small part of the feed spacer or the membrane, and in addition it cannot be used to measure fouling processes directly *in-situ* [102-104]. Fouling characterization has also been carried out using electrical impedance spectroscopy [105,106] and ultrasonic time domain reflectometry [107,108] , but again it is difficult for these sensor-based methods to produce high resolution images of the fouling process directly.

#### **3.2 Tomography Based Techniques**

Optical coherence tomography (OCT) produces a three-dimensional image of the internal structure of a material at micrometre resolution from the detection of light reflected by internal features, in a process analogous to reconstruction of structure from ultrasound data [109,110]. In Fourier (or frequency) Domain OCT, an interference fringe signal is initially gained as a function of optical frequency by combining the back-reflected signal from the sample with that obtained from a reference with a known delay. The phase angle and magnitude of the signals are re-formed in the spatial domain through a Fourier transform[82]. OCT is a non-destructive and *in-situ* technique that can be employed to characterise the flow patterns through the feed spacer in a membrane filtration module and can gain detailed structure of the biofouling layer [111,112]. Recently, the adoption of OCT to study biomass accumulation behaviour in membrane systems has increased. OCT has been used to study biofilm formation and evolution on aqueous surfaces due to its ability to allow measurements

of biofilm formation *in-situ* and without additional preparation required [109]. The ability to observe a specific section of the feed spacer in the mm range is an advantageous feature of OCT, and particularly significant for the better understanding of biofouling growth, where the feed spacer can be completely examined [113,114]. Optical coherence tomography has also been employed analyses in order to calculate biofilm thickness and to evaluate the performance of two-phase cleaning fluid flow in a spiral-wound element [115,116]. A major advantage of the application of the OCT system is characterisation of the fluid dynamics during filtration process[111], as well as reliable acquisition for three-dimensional data of the fouling deposit in the membrane system[102].

Gao *et al.*[102] obtained a series of OCT images as a function of filtration time with different fluid flow conditions, including with and without a feed spacer in the fluid channel (see Figure 14) [111]. Fouling during membrane filtration was observed, with the effect of feed spacer orientation being of particular interest. Transport phenomena through the feed spacer interstices were characterized from the tomographic images obtained. Images of the fluid velocity profile in the channel filled with a feed spacer inclined at 45° to the direction of the bulk flow can be seen in Figure 15. Wibisono *et al.* [116] employed OCT to image biofouling layer removal through two-phase fluid flow in a nano-filtration membrane system in the presence of a net spacer. They stated that the advantage of adopting the OCT method for visualization of biofilms is that image acquisition was possible without a need to open the cultivation chamber, allowing *in-situ* determination of cross-sections of the biofilm deposits at the membrane surface. OCT for 3D *in situ* observations of feed spacer channels was applied by West *et al* [114] to quantify the biofouling coverage in a membrane fouling simulator, set to mimic the spacer channel of spiral wound membrane modules, without disturbing the filtration process. Effluent of a water treatment unit and river water were employed as feed solutions to visualize feed spacer fouling behaviour. The influence of the spacer geometry on the fouling accumulation was assessed for both a wide-net and small-net feed spacer, to explore the potential of OCT for the analysis of membrane fouling. They stated that OCT is a feasible device to monitor biofouling layer, which supplies high resolution three-dimensional information about the biofilm deposit without any drawbacks (as shown in Figure 16).

Haaksman *et al* [69] utilized X-ray computed tomography to investigate the cross-sectional size and shape of commercial feed spacer filaments and used this information to obtain an accurate dataset of feed spacer geometries and to develop computational fluid dynamics (CDF) models. Local velocity and shear distribution were quantified, leading to improved estimations of concentration polarization and fouling accumulation (see Figure 17). Results of the CDF models were in closer agreement with experimental data than previous models which assumed a simpler filament geometry. As a result, computed tomography scan data of spacer filament geometry allows models with more accurate estimation of local velocity and shear rates, leading to improved spacer design. The authors used this information to develop a spacer with novel geometry which had a decreased pressure drop and high shear rates at the membrane surface.

Another non-destructive quantification of biomass creation at a spacer under typical conditions for membrane systems using OCT was reported by Fortunato *et al* [117], in a membrane fouling simulator (flow channel containing membrane and feed spacer). When the fluid-flow cell was monitored for five days using an ultrafiltration membrane, pressure drop and permeate flux was observed. It allows *in-situ* biofilm evaluation in a flow channel with both the feed spacer and membrane, wherein the detection of biofouling was detected by an OCT device (see Figure 18). Fortunato and Leiknes [113] utilised three-dimensional OCT image analysis in order to evaluate *in-situ* spatial-resolved and time-resolved imaging of the

biofouling layer accumulation pattern. This approach was presented using an image analysis procedure to gain biofilm thickness maps used to measure biofouling formation and distribution in membrane systems over time. The maps allow evaluation of the biofilm growth through use of a false color scale (see Figure 19) and can also be beneficial in evaluating antifouling strategies. The method can be used to characterize the biofilm and can also be helpful in evaluating the efficiency of fouling mitigation and de-fouling strategies. Feed spacer size and shape have effects on hydrodynamics of membrane systems as well as performance of the system, inaccurate measurements may lead to poor predictions of hydrodynamics and membrane performance. Therefore, accurate measurement is required to understand and enhance the fluid dynamics models used for membrane module design. Siddiqui *et al* [82] applied six methods to measure the feed spacer porosity, cuboidal, ellipsoidal and cylindrical volume calculation, volume displacement, weight and density and computed tomography scanning. The six-adopted mesh-spacers examined varied in mesh-size, the angle between the filaments and filament thickness. There was disagreement in porosity measured by the different measurement techniques, and it was found that greater accuracy was found for scanning, weight and density and volume displacement.

#### **4. Conclusion**

Currently, the adoption of feed spacers with high performance in membrane systems designed for fouling mitigation shows great potential in water treatment technology. It is too early to conclude whether spacer modification has now reached a state of maturity, as numerous possibilities for improvement still exist. Efforts should continue towards the enhancement of high flux and low-pressure drop in membrane modules with improved resistance to fouling. Further attention should be paid on the relation between modification of the feed spacer and the underlying membrane performance. This domain needs intensive analyses as there are no comprehensive studies into what type of idealized feed spacer modification (e.g. surface coating, shape geometry, conductivity, etc.) could match improved membrane performance for particular industrial applications. It is hoped that continuous modification of feed spacer configuration and applications with respect to enhancement of membrane permeability and selectivity in the future will increase the performance of membrane module to provide excellent selectivity without a sacrifice in water productivity. Much work has been done on finding optimum configurations of feed spacer geometry to minimise fouling by improving mixing to reduce concentration polarisation and to increase shear forces at the membrane surface. In addition methods to employ electrolytic in situ cleaning of membrane modules using electrically conducting feed spacers are very promising. So far much work has been done in the lab scale and with computer modelling. The major challenge now is to perfect these advances and scale them up for incorporation into commercially available systems.

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