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The evolution of feed spacer role in membrane applications for desalination and water treatment: A critical review and future perspective

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HIGHLIGHTS

- Research progress in feed spacer development and enhancement is presented.
- Quantitative and qualitative tools for spacer performance assessment are discussed.
- Spacer improvement discussed based on novel geometries and surface modification.
- Future perspective and emerging applications of feed spacers are highlighted.
- Role of functionalized spacers, 4D printing and smart materials is emphasized.

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ABSTRACT

The membrane research community has witnessed a significant research growth on feed spacers since 2010, both in quantity and scope. Increasingly novel spacer geometries and spacer chemistries are being reported in literature to tackle key challenges facing membrane processes, including concentration polarization, energy consumption, low flux, and fouling. This review aims to analyze the recent developments of spacer research, starting with a review of the various roles spacers can and do play in membrane systems and their increased prominence in tackling membrane fouling and scaling. We then review the continued need for novel spacer designs, discuss the contribution spacers can make to energy saving, and the necessity for new generations of spacers to facilitate the deployment of next generation, ultra-permeable membranes. Qualitative and quantitative spacer performance assessment tools are described. The key trends in spacer modifications over the period of 2010 to 2021 are categorized into: i) design modifications of both conventional and novel geometries, and ii) spacer surface modifications, including chemical coatings and new spacer materials. Finally, the future trends of spacer research are highlighted, with an emphasis on the development of functionalized feed spacers and new spacers made via 4D printing and smart materials.

1. Introduction

Desalination, water treatment, and wastewater reclamation, specifically via membrane technology, are considered to be key solutions to the global water crisis exacerbated by worsening impacts of climate change, increased urbanization, population growth and industrialization. The membranes in various separation technologies are typically housed in one of four types of modules: plate and frame, tubular, hollow-fiber, and spiral wound [1]. The growth of membrane processes, in particular reverse osmosis (RO), has coincided with the rise of the spiral wound module (SWM) over the hollow fiber configuration [2]. The feed spacer, an essential component of the SWMs, is also found in plate and frame and -potentially- in tubular modules as static turbulence promoter. A typical feed spacer in an SWM is shown in Fig. 1(a). The feed spacer netting is placed in between the membrane sheets, facing the active layer of the membranes, while a permeate spacer (called "permeate carrier") is placed on the permeate side. The permeate is collected in the perforated tube placed in the center of the module [3]. The spacer essentially performs two roles within the module: i) to provide separation between the membrane leaves and to support them; and

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ii) to enhance flow mixing and increase unsteadiness within the feed channel, in order to increase mass transfer in order to reduce concentration polarization (CP). The conventional feed spacer is usually made from polypropylene, due to its chemical inertness, ease to fabricate by extrusion, and low cost [2].

The conventional design of commercial net-type spacer, which is most commonly found in SWMs today, consists of two layers of cylindrical filaments arranged over each other so as to form a parallelogram, where l_m and d_f are the length and diameter of the filament within each block of the mesh, respectively (Fig. 1(b)). Depending on the spacer design, l_m and d_f may be the same for each filament in the spacer, or different, as in the case of alternating strand design (ASD) by LANXESS AD [4]. In commercial SWM, the two layers of filaments are in contact with the two membrane active surfaces. The angle at which the two layers of filaments intersect is called β, which typically varies from 60° to 120°, and the angle at which the feed approaches the spacer is called the flow attack angle, α [5]. While the feed spacer is expected to enhance unsteady flow and minimize CP in the feed channel, via enhancing the mass transfer coefficient, there is a well-known trade-off with respect to increased pressure drop [6]. Regardless, the flux enhancement provided by the spacer has made this trade-off acceptable. Many novel spacer designs, as will be discussed thoroughly in this review, essentially aim to shift the balance of this trade-off further in favor of increased mass transfer.

Fig. 2 depicts a timeline for major industrial and research developments pertaining to feed spacers. The consolidation of two leading spacer manufacturers under a single conglomerate is notable, as Schweitzer-Mauduit International (also known as SWM) acquired the two major companies in spacer production, Delstar and Conwed Plastics in 2013 and 2017, respectively [9]. Today, the new SWM company controls more than 90 % of the global spacer market and has identified, on its website, innovative designs as a key development area for future spacers. 3D printing of feed spacer was first reported in 2005 [10], though it would take almost a decade for a notable proliferation of research in this direction, especially on novel spacer designs. In 2009, a significant research paper was published by Vrouwenvelder et al. that identified feed spacers as the chief culprits in increased feed channel pressure drop (FCPD) due to biofouling [11], which redirected spacer research significantly in the following years.

The growth trend in feed spacer research over the period of 2010 to 2021 is reflected in a clear annual growth of publications, as per data collected and analyzed from Scopus using the keywords (membrane +

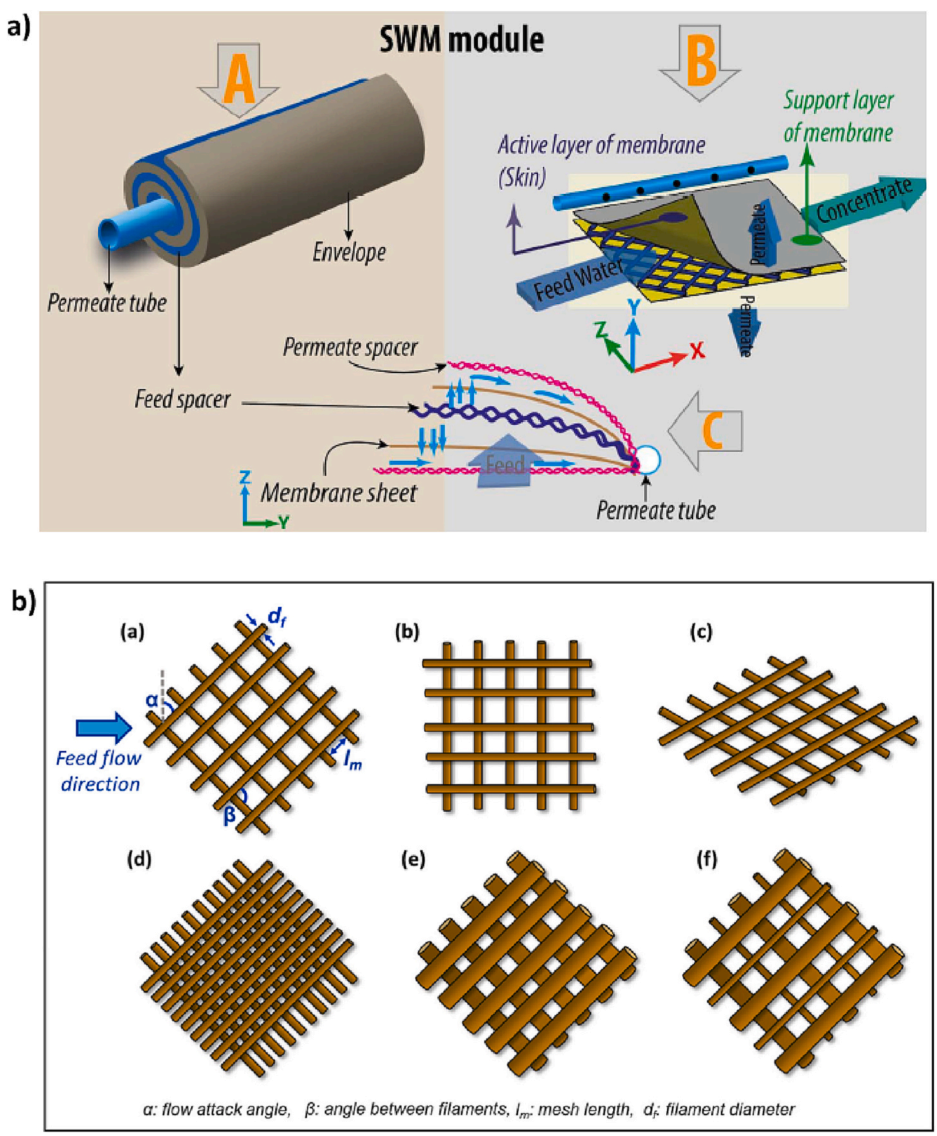

Fig. 1. a) Illustration of a typical spiral wound module (SWM) and its components [7]; b) net-type commercial spacers and design variations based on the different spacer parameters [8].

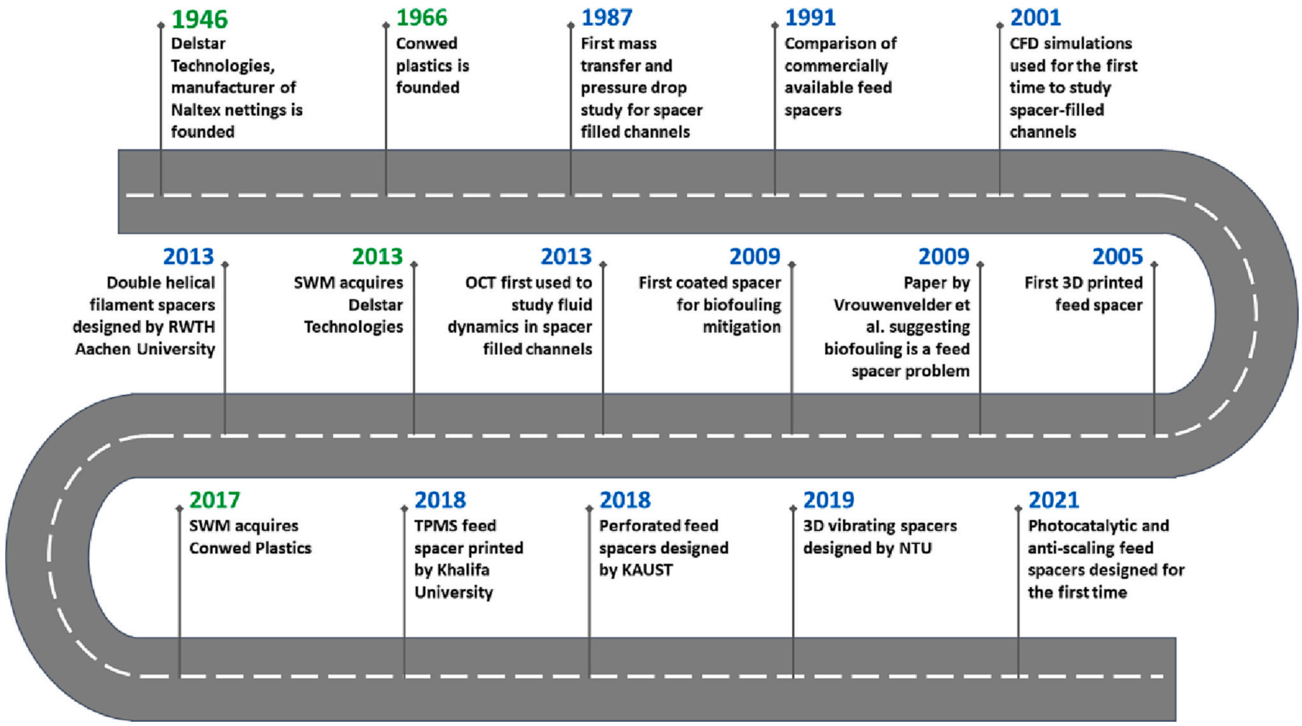

Fig. 2. Timeline of major feed spacer developments in industry (marked in green font) and academia (marked in blue font) since 1946.

feed spacer) (Fig. 3(a)). This rise in publications has coincided with a growing number of papers on the development and testing of spacer with novel designs. An interesting finding was that, despite the fact that constant flux is more common in commercial operations [12], far more spacer research was conducted under constant pressure operating mode than constant flux operation. Fig. 3(b) shows the breakdown of spacer studies based on studied membrane application. RO, ultrafiltration (UF), and microfiltration (MF) were the most studied processes for testing spacers. It is also interesting to note that emerging membrane processes, such as forward osmosis (FO), membrane distillation (MD), and pressure retarded osmosis (PRO), are gaining prominence in spacer research recently, with spacers being tested more frequently in these processes for fouling and scaling reduction, as discussed later.

1.1. The need for this review

There has been a number of reviews in recent years that tangentially addressed feed spacers in the context of: i) 3D printing in membrane technology [13–19], ii) fouling mitigation [20], iii) membrane biofouling [21], iv) summarizing early spacer research (until 2004) [22], or v) summarizing spacer research in recent years [23]. However, there has been minimal in-depth discussion in the said reviews on why exactly novel spacers are needed or the direction in which spacer research is headed. To fill this gap, this review aims to i) illustrate the

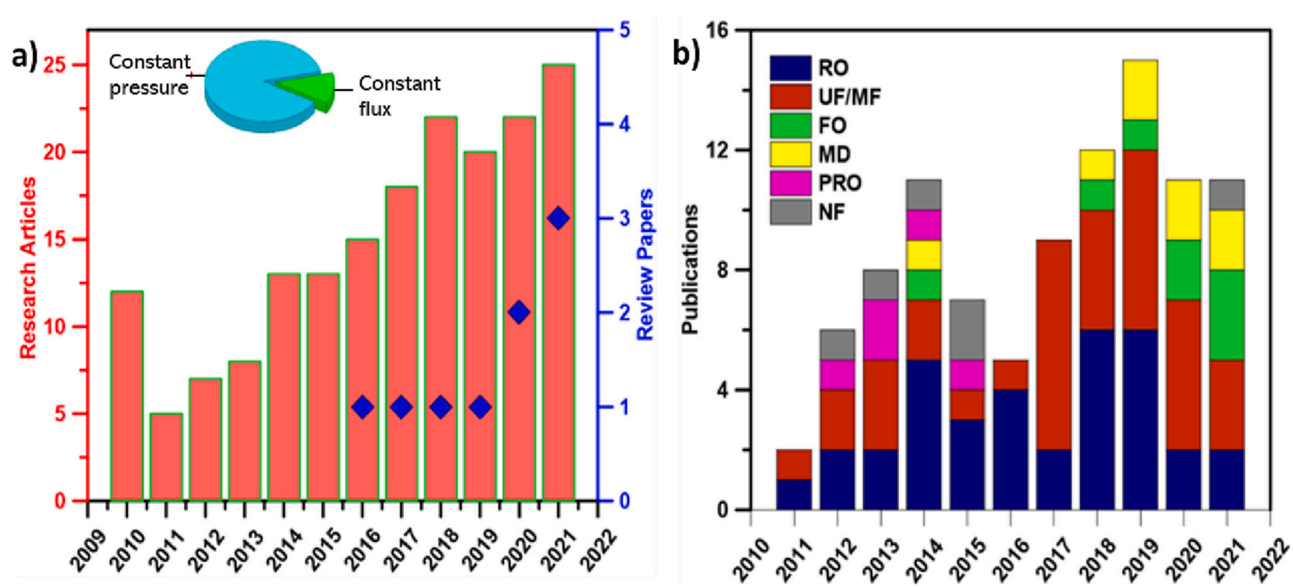

Fig. 3. a) Growth in spacer research over the period of 2010 to 2021 with red bars representing research articles and blue points representing review papers (inset: ratio of studies conducted in constant pressure mode versus constant flux mode); b) breakdown of spacer studies based on studied membrane application.

tools that have been used to study spacers and their performance, ii) detail spacer research as it has been redefined in the last 10 years, and iii) provide an educated opinion on the direction spacer research can and should take in the coming years. Importantly, this review aims to re-contextualize spacer research in terms of where membrane and water research is heading in general, to emphasize the necessity of developing more innovative spacers, and to present the potential avenues from which profound advances in this field could be achieved.

To the best of the authors' knowledge, this is the first review that discusses in detail the need for spacer research and development for the future of membrane technology in water filtration and desalination applications. The discussion is developed in terms of tackling the menace of fouling in membrane systems, the minimization of energy losses in various processes, as well as the optimization of novel, high performance membranes being developed in literature. The various tools that can be used to analyze spacer performances are illustrated, while discussing the potential for large-scale application where appropriate. It should be noted that most of the processes discussed in this review pertain to desalination either directly (RO, MD) or indirectly (UF, NF etc.). The relevance of feed spacers is also significantly tied to the SWM which is prominent in desalination plants. Nonetheless, numerous studies with feed spacers have employed feed waters that are analogous to industrial and biological wastewaters. This is relevant due to the increased focus of spacers in biofouling and scaling, which are readily assessed with these types of feeds. It is also an indication of the growing applications of membranes in other water treatment processes, which subsequently increases the relevance of feed spacer research to treat various feedwaters. This is meant to be a comprehensive review that attempts to provide a holistic overview of spacer research in recent past and in coming years.

This review has been organized under 4 sections. Section 1 introduces spacers and the role they play in membrane processes, with Section 1.1 advocating for the need for this review, Section 1.2 describing the role spacers play in different types of fouling, and Section 1.3 explaining the need for advanced spacer designs (readers are referred to other recent publications [7,22,23] that has detailed spacer-based performance enhancements in membrane applications). Section 2 categorizes the various tools that have been used to benchmark spacer performances, which include quantitative (Section 2.1) and qualitative (Section 2.2) tools and a combination thereof (Section 2.3). It also highlights the concerns regarding some of these tools vis-à-vis the scale at which observations are made (Section 2.4). Section 3 describes the different aspects of spacer research over the period of 2010 to 2021, with a focus on surface modifications of spacers (Section 3.1) and the modification of spacer geometries (Section 3.2). Finally, Section 4 forecasts the next frontier of spacer research and includes discussion on the challenges of scaling up production of novel spacers (Section 4.1), development of novel functionalized feed spacers (Section 4.2), as well as the incorporation of 4D printing and smart materials into spacer design (Section 4.3).

1.2. Spacers and fouling

While spacers in membrane systems have been designed to reduce CP and increase mass transfer through internal mixing, they have a significant influence on another major aspect of membrane processes; fouling. Fouling refers to the deposition and/or build-up of unwanted materials on the membrane or spacer, which subsequently affects the performance of the process and/or permeate quality. This remains a major hurdle in the increased acceptance of membrane processes globally [24,25]. Fouling increases process costs in terms of energy requirements, cleaning chemicals, and membrane replacements [26]. Fouling also has an associated environmental cost, in the form of increased greenhouse gas emissions, higher chemical discharges in reject stream, and a build-up of spent membrane modules [27,28]. A wide variety of particles, colloids, and dissolved materials, such as metal oxides, dissolved scaling salts, microbes, and macromolecules, can be considered foulants.

Membrane fouling is categorized into four main categories: organic fouling, colloidal fouling, biofouling, and inorganic fouling (i.e., scaling) [21]. Historically, biofouling received significant attention as it is believed to be the most detrimental form of membrane fouling [21]. Biofouling is a difficult phenomenon to eliminate completely, considering that even with pretreatment processes, the complete elimination of microbes is difficult to achieve. Conventional pretreatment methods such as sedimentation and coagulation/flocculation can remove most suspended particles, and the addition of antiscalants can reduce the deposition of scale-forming salts. However, microorganisms are capable of growing, multiplying and relocating on a membrane surface, making them significantly more complex to deal with [25,29]. A small percentage of microbes can subsequently develop into biofilms that are extremely resilient and tough to remove, even with continuous use of cleaning chemicals [30]. Additionally, incorrect dosages of antiscalants can lead to greater microbial growth as they can provide an additional nutrient source for the biofilm community, while traditional anti-bacterial technologies such as chlorination and ozonation can develop harmful chemical by-products in the feed and damage RO membranes [25]. Therefore, significant effort was dedicated to developing anti-biofouling strategies. And, since enhancing the pretreatment has no guarantee of success, particularly for unpredictable and challenging feed sources, the focus has been mainly on modifying membrane properties such as surface charge, hydrophilicity and surface roughness, or the incorporation of biocidal materials into the membrane polymer or on its surface [11,29].

The fact that feed spacers have an effect on biofilm development within the membrane module has been known for sometime. In 1995, Baker et al. noted initial deposition of foulants along spacer filaments, which subsequently encroached onto the membrane area [31], and, in 1998, van Paassen et al. noted the increase in FCPD due to biofouling on the feed spacers [32]. In 1997, Ridgway suggested that improved hydrodynamics through spacer design might be the solution to biofouling [33]. In 2007, Tran et al. also noted an increased occurrence of fouling next to spacer filaments [34]. In spite of these corroborating observations, membrane modification remained the focus of biofouling mitigation studies during this period. However, a series of studies led to an increased attention to feed spacers as a pathway to target biofouling. Vrouwenvelder et al. found that feed spacers had the most significant effect on the negative outcomes of biofouling, such as FCPD, while other factors such as membrane type, transmembrane pressure and even permeate flux had a limited effect (Fig. 4(a)) [11,35]. They conducted 3D computational fluid dynamics modeling to re-establish that dead zone regions (where feed spacers block the feed flow) are the most susceptible to microbial attachment and subsequent biofilm growth (Fig. 4(b)) [36,37]. They also conclusively established a link between net-type spacer design parameters such as thickness, porosity, filament shape and thickness, and the FCPD from biofouling [38]. Further research concluded that particle deposition in general is affected by spacer geometry, and that spacer modification should be a strategy for fouling mitigation [39]. Nevertheless, several studies emerged to counter this view. Suwarno et al. conducted a study of biofouling over a range of transmembrane pressure (TMP) and spacer variations to validate the conclusions of Vrouvenwelder et al. The results were mixed and Suwarno et al. postulated that, depending on the conditions of operation, membrane fouling or spacer fouling may be more dominant, although the study also noted the correlation between spacer design and biofouling (Fig. 4(c)) [40]. A more recent study by Lin et al. confirmed, once again, that spacer filaments' intersections were the points of origin for biofouling and that the geometrical parameters of spacers have significant effects on the latter [41]. It is safe to conclude that spacers certainly play a role in biofouling and have a significant effect on the FCPD resulting from it. It is also reasonable to assume that regions with low hydrodynamic mixing will be most conducive to microbial attachment. The differing conclusions of the studies conducted globally can be

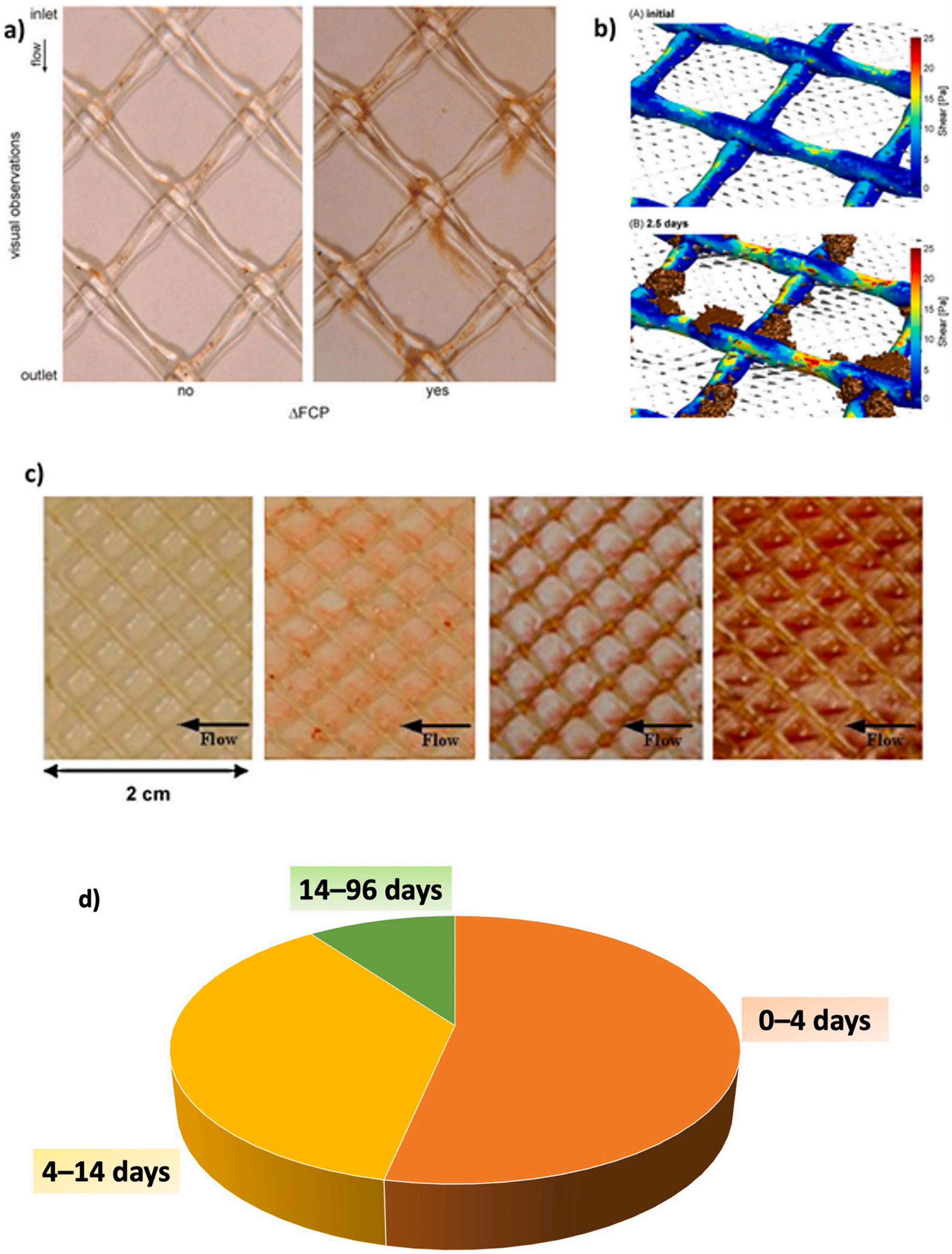

Fig. 4. Examples from literature indicating a correlation between spacer use and membrane fouling. a) In situ observation of fouling in membrane cell with substrate dosage (right) and without substrate dosage (left) [11]; b) biofilm development as seen using CFD modeling after 0 days (top) and 2.5 days (bottom) [36]; c) photographs of biofouling as it evolves over a period of 10 days, from left to right [40]. d) distribution of experimental duration for 30 studies with feed spacers in biofouling studies.

explained based on their differences in experimental conditions, including test units variations, membranes types used, and feed characteristics. It is also worth pondering that the differences may lie in the interpretation of results, which may not always be contradictory. For instance, while Vrouwenvelder et al. often suggest that FCPD due to biofouling is higher with spacers, it may not necessarily mean that spacer fouling is higher than membrane fouling, but merely that the impact of spacers is significant. Suwarno et al. also noted differences in biofouling with and without feed spacer, and in the absence of spacer a much higher TMP was observed, while the difference in biomass was minimal [40]. Bacterial colonies developed along the membrane feed channel in a uniform pattern, rather than clustering around spacer filaments, and were eventually covered by a thick layer of dead bacteria. A potential factor determining membrane fouling over spacer fouling could be the nutrient load in the feed. When this load is low, the amount of shear forces generated by the spacer can prevent excessive concentration of nutrients on the boundary layer by increased mixing, hence reduced biofouling on the membrane, while the regions next to the spacer filament have adequate nutrients for biofilm development. When nutrients are plentiful, there is increased concentration polarization on the boundary layer, leading to suitable conditions on the membrane for biofouling. In this case, the spacer impact may be less relevant due to the abundance of nutrients, whereas membrane properties such as surface energy and roughness become more relevant, and membrane dominated biofouling may occur. Further studies are necessary to gain a deeper understanding of the relative roles of membranes and spacers in biofouling. Another aspect to consider, particularly with biofouling studies, is the experimental duration that these studies are conducted for (Fig. 4(d)). It has previously been demonstrated that short-term biofouling tests comparing spacer modifications might not be extrapolated to similar results during long term tests [42]. However, we can see that most studies are conducted for shorter time frames. In order to accurately understand the effect of spacers on biofilms in commercial modules, longer experiments mimicking plant operation conditions are needed.

There has generally been a shortage of studies that investigated the effects of feed spacers on membrane scaling, although few studies evaluated the effect of feed spacers on mass and heat transfer and temperature polarization (TP) in MD [43–45]. Given that scaling is largely a function of supersaturation at the membrane surface, which leads to nucleation of salt crystals, it was logical to assume that spacers could at least indirectly affect the scaling in the system due to its impact on localized mixing [5]. Thomas et al. studied the effect of novel spacers designed based on triply periodic minimal surfaces (TPMS) on calcium sulphate scale development in direct contact MD [46]. These authors studied the performance of functionalized 3D printed feed spacers as well, by coating fluorinated silica to probe its impact of altering the surface energy on scaling [47]. The coated spacers showed significant reduction in calcium sulphate scaling on both the spacer (reduced by 74 %) and the membrane (reduced by 60 %). Recent work by Anvari et al. on stainless steel spacers heated by radio frequency showed promise in enhancing flux and reducing scaling in MD, though further studies are still needed to support this approach [48]. An electrically polarized graphene polylactic acid spacer developed by Yanar et al. was able to demonstrate a reduction in gypsum scaling in FO [49]. Overall, the potential to reduce scaling using spacer designs and spacer coatings in a variety of applications shows promise, and should be pursued more rigorously in the future.

1.3. Need for advanced spacers

While the majority of spacer research since 2010 focused on addressing fouling, the impact that spacers have on feed channel flow regime, unsteady flow and CP remains a significant driver for spacer research in general and for novel spacer designs development in particular. Firstly, consider the current membrane module design and energy consumption, taking the example of desalination through the spiral wound module design. The minimum theoretical separation energy for seawater (35,000 ppm) desalination under ideal conditions and 50 % recovery is calculated to be 1.06 kWh/m^3. The actual energy requirement is higher, as desalination is not an ideal (i.e., reversible) process [50]. While current state of the art seawater RO technology can operate at an energy requirement of 3–4 kWh/m^3, a target of 1.5–2 kWh/m^3 is considered realistic in the future [51]. It is worth noting that while innovations in module design, low energy membranes, pump design, and energy recovery devices have already brought down the energy requirements of RO significantly, energy still accounts for around 50 % of the total seawater desalination cost [52]. In an analysis conducted by Shrivastava et al., the additional energy required for the RO process (beyond the theoretical minimum required for salt-water separation) was termed W_{RO}. W_{RO} is a summation of energy losses in the module (E_{Module}), energy losses of the membrane (E_{Flow}) and energy losses in the system (E_{System}). While E_{Flow} and E_{System} may have already been minimized, due to advancements in technologies such as high efficiency pumps and energy recovery devices, there is still potential to reduce E_{Module} [52]. E_{Module} largely refers to energy losses due to friction in the feed channel, as well as CP, both of which are influenced by the feed spacer. Therefore, improved spacer design is a realistic avenue to reduce energy consumption in membrane processes.

There is yet another critical reason why improved spacer designs are needed. A major frontier in water research today is the development of novel membranes that can overcome the limitations of current crop of membrane materials. Thin-film composite polyamide membranes have driven the growth of RO technology for four decades but have seemingly approached their limits with respect to permeability-selectivity trade-off. They are also hindered by their sensitivity to chlorine, which in turn makes biofouling an ever-present issue [53,54]. Consequently, many researchers have been attempting to develop a new generation of membranes with significantly greater permeability (called ultra-permeable membranes, UPMs), without compromising membrane selectivity [55]. UPMs are designed using a variety of materials and techniques, from carbon-nanotubes and graphene to biomimetic membranes [56]. While these UPMs are expected to have significantly higher permeabilities, in practice, they will still be limited by CP when scaled-up. McGovern et al. [57] noted that there will be a limiting flux for UPMs, which is dependent on their mass transfer coefficient. And, even though there are expected advantages to UPMs, those may not be as significant as originally envisioned (see Fig. 5(a)) [58]. With this in mind, it will be necessary to develop advanced feed spacers that can drive up the mass transfer coefficient in order to take advantage of these UPMs [59]. Toh et al. conducted a techno-economic analysis where they studied the impact of combining an advanced spacer with high-permeance membranes. Their analysis concluded that advanced spacers can provide significant advantages in increasing permeance, and there is a strong case to be made for the development of novel spacers that enhance mass transfer coefficient and minimize energy losses (see Fig. 5(b)).

2. Performance evaluation of feed spacers

Due to the wide range of spacer designs, modifications, and applications which they have been used in, a wide variety of tools has been employed to evaluate their performance. These tools, which can be qualitative, quantitative, or a combination thereof, are critical in assessing the spacer's efficiency and in benchmarking new spacer designs. Fig. 6 demonstrates a relative distribution of all spacer evaluation tools, which were reported in all spacer research papers published in the period from 2010 to 2021. These tools range from performance-related parameters, like flux and FCPD, to visual observations of fouling. The following subsections discuss the most commonly used tools.

2.1. Quantitative tools

2.1.1. Flux/TMP change

The effect of spacers on flux or TMP (depending on the operating mode) is one of the simplest and most widely utilized analyses that can be conducted in a test setup. It is worth noting here that most early spacer studies were conducted in constant TMP mode. As noted earlier, constant flux is the more common operating mode commercially, however, it is traditionally harder to operate under lab conditions. Numerous examples are seen of flux and TMP being used to evaluate spacer performance [6,61]. The study performed by Fritzmann et al. to evaluate their microstructured twisting double-helix spacers is one of the early examples of TMP rise usage to characterize spacer performances [62]. Sreedhar et al. utilized permeate flux to evaluate the TPMS spacers initially [63], although their later studies also evaluated spacer efficiency based on changes in TMP [64]. For MD, flux has typically been used in evaluating spacer performance vis-à-vis mass and heat transfer enhancement, as well as scaling mitigation. Thomas et al. utilized flux to evaluate the performance of TPMS spacers in multiple MD studies (see Fig. 7(a)) [46,65,66]. Kerdi et al. employed permeate flux to evaluate their perforated [67] and helical filament [68] spacers. Ali et al. reported that specific flux, a parameter which incorporates both FCPD and flux, can be a better tool to evaluate spacer efficiency, as demonstrated for their dynamic turbospacer [69].

2.1.2. Feed channel pressure drop (FCPD)

FCPD is the difference between the feed and the reject pressures, and is typically directly impacted by the spacer design due **to** flow obstruction induced by the latter [23]. In the presence of a feed spacer, biofouling-induced FCPD can become more pronounced, as reported by Vrouwenvelder et al. [11]. A number of studies employed FCPD to

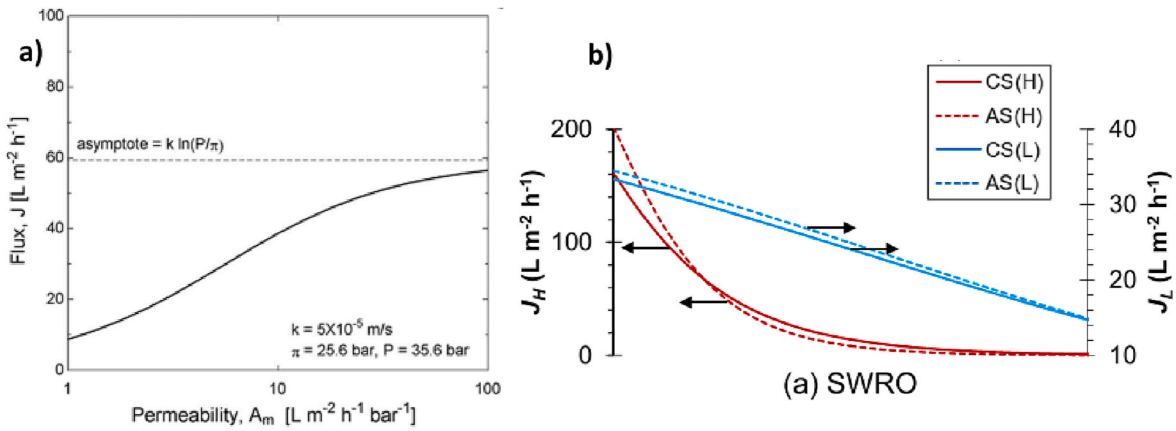

Fig. 5. a) Flux versus recovery plot showing an asymptote flux for seawater RO [57]; b) permeate flux along the length of the membrane for conventional spacer (CS), advanced spacer (AS), using low (L) and high (H) permeability membranes [60].

analyze the effect of fouling, as well as to characterize the hydraulic efficiency of spacers in minimizing energy losses (see Fig. 7(b)). Wibisono et al. reported the change in FCPD to evaluate their chemically coated feed spacers [70], while Dreszer et al. measured FCPD to determine the effects of spacers, nutrients and crossflow velocity on fouling [71]. Other researchers, such as Jablonska et al., used FCPD to evaluate the efficiency of their coated spacers as well [72]. Lin et al. and Sreedhar et al. used FCPD in the absence of fouling to characterize the hydraulic resistance of net-type and TPMS feed spacer designs, respectively [41,65]. Kerdi et al. also used FCPD to characterize their perforated spacers in a similar manner [67]. While FCPD provides a simple way to efficiently characterize the hydraulic performance of spacers, as an assessment tool, it can suffer from non-uniformity of test methods, sensors used, and module design, all of which can affect the FCPD measurements made in the lab.

2.1.3. Dimensionless analysis

A number of researchers have utilized dimensionless numbers, such as Reynolds number (*Re*), Sherwood number (*Sh*), Power number (*Pn*), Schmidt number (*Sc*), and friction factor, to quantitatively assess spacer performance. In the early years of spacer research, Da Costa et al. carried out studies to compare net-type spacers available at that time using dimensionless numbers [75–78]. In the early 2000s, Li et al. employed *Sh* to evaluate spacer designs [10,79], as was later done by Koutsou et al. and Fimbres-Weihs et al. [80,81]. More recently, dimensionless numbers were employed by Tan et al. to compare net-type spacers printed using different additive manufacturing techniques (see Fig. 7(c)) [74], and by Sreedhar et al. to compare TPMS spacers with changing thicknesses in UF [82]. This tool was also employed by Ruiz-Garcia et al. to study the effect of spacer geometries and membrane permeabilities on brackish water RO (BWRO) [83]. Dimensionless analysis potentially offers a reliable method to study spacers while neutralizing the effects of other variables, such as operating conditions, module geometry, etc. This information can be very valuable in the development of correlations that allow the development of modules with novel spacers, providing a framework for the scale-up and design of these new modules. However, this tool has not been regularly employed in spacer research. One reason for this is the need to employ simple feed with well-known properties, such as Dextran T-500, which was utilized by Da Costa et al., Tan et al., and Sreedhar et al. [74,75,82]. The need to calculate the density, viscosity and osmotic pressure of the feed used to determine the mass transfer coefficient of the spacer severely limits the use of dimensionless numbers in filtration studies to single solute feeds, for which these properties have been well-established in literature. In a recent study by Sreedhar et al. it was noted that different feed solutions can lead to different degrees of fouling and cake formation on the membrane, with a comparison carried out between Dextran and sodium alginate [84]. It is also worth noting that within a commercial module, fluid conditions are changing constantly across the length of the module, as well as from one module to another, and these variations are not accounted for in dimensionless analysis.

2.1.4. Fouling and cake resistance models

The development of cake filtration model and its variations for reversible and irreversible fouling has allowed the quantification of foulant cake layer properties, and was therefore employed in studying fouling mitigation strategies [85,86]. Recently, these models started to be employed in studying novel spacers as well. Sreedhar et al. studied reversible and irreversible fouling layers in the cake formed during sodium alginate filtration using different spacers and compared their effects on membrane cleaning operations, such as backwash and relaxation [64]. Similarly, cake resistance was determined by Park et al. to benchmark their 3D printed honeycomb spacers against commercial spacers [87]. Fouling layer resistance was also used by Ali et al. to study their dynamic turbospacers in FO [88] and was utilized by Ghaffour and Qamar to study helical cleaners for tubular modules [89].

2.1.5. Total organic carbon (TOC) and adenosine triphosphate (ATP)

TOC and ATP are utilized as means of quantifying the amount of biofilm on the membrane in biofouling studies (Fig. 7(d)). While TOC provides an estimate of the biomass on the membrane, including both microbial cells and extra-cellular polymeric substances (EPS), ATP gives an estimate of living cells in the biofilm. Both have been employed in numerous studies such as the study by Araujo et al. in 2012 on anti-adhesive and antibacterial coatings, as well as antiadhesive coatings for both membranes and spacers by Miller et al. [42,90]. Linares et al. utilized ATP to study the effects of spacer thickness on biofouling in FO [91] and Ronen et al. used TOC to study membrane area fouled after using feed spacers coated by quaternary ammonium groups and silver nanoparticles [92]. Siddiqui et al. utilized both TOC and ATP to study 3D printed net-type spacers for biofouling with tap water [93] and Sreedhar et al. utilized TOC to study membrane fouling using TPMS spacers and net-type spacers [63]. Bogler et al. utilized TOC to study membrane biofouling using a super hydrophilic zwitterionic-coated feed spacer [94].

While TOC and ATP provide a swift and simple method to analyze biofouling, they must be used with caution. Although TOC provides an insight on the amount of organic matter on the membrane, in complex feed systems, such as river water and industrial waste, the amount of TOC cannot be directly equated to biofilm, as other organic substances in the feed can contribute to the measured TOC. Likewise, while ATP provides an estimate of living cells, this cannot provide a holistic

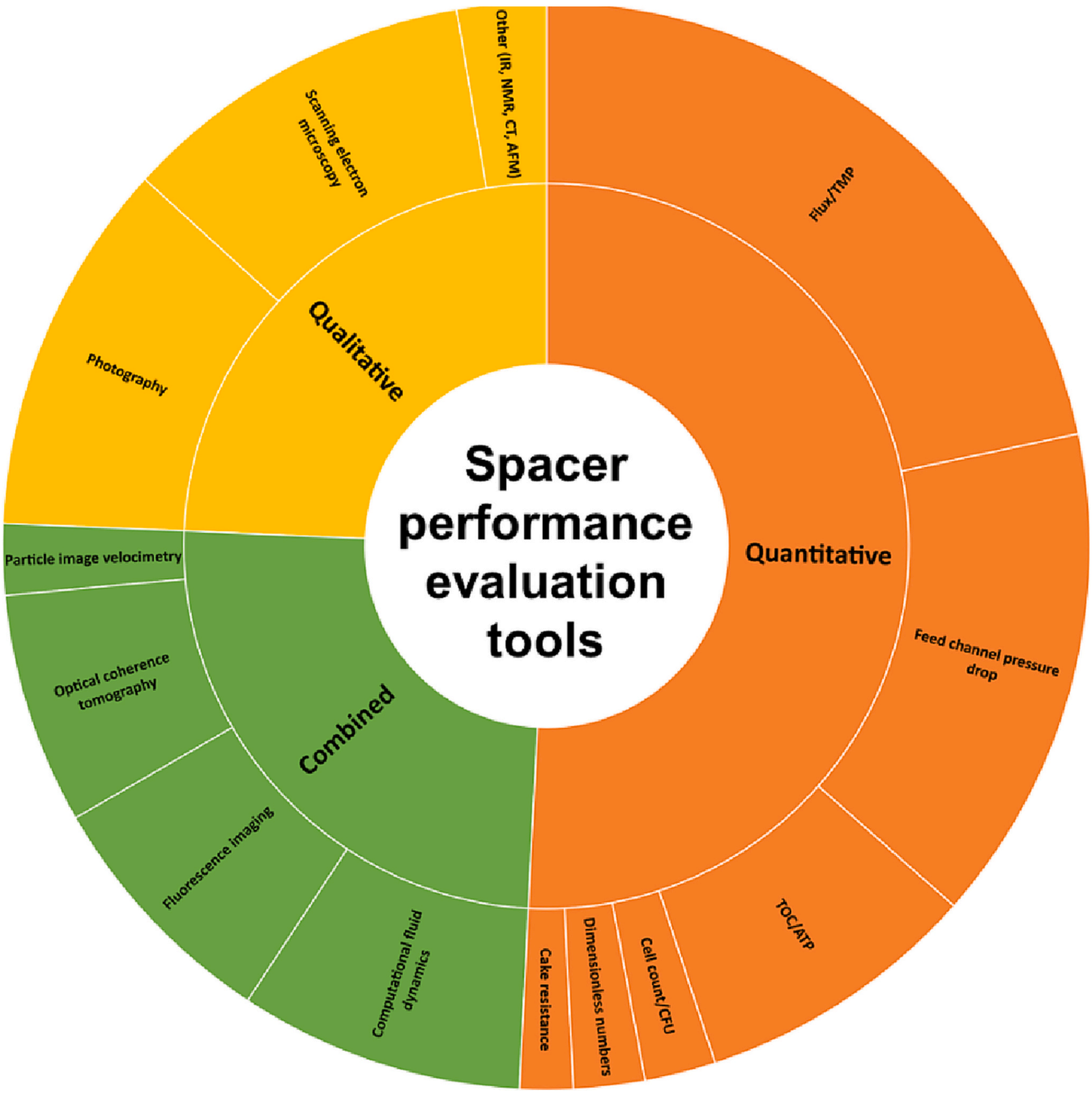

Fig. 6. A relative distribution of the tools used in spacer performance evaluation from 2010 to 2021. The tools are sub-categorized to qualitative, quantitative and a combination thereof, with several tools under each category.

analysis, as dead cells are likely to constitute part of the biofilm as well [95,96].

2.2. Qualitative tools

2.2.1. SEM imaging

Scanning electron microscopy (SEM) is often employed to observe the membrane or spacer after fouling has taken place (Fig. 8(a)). For example, Hausman et al. viewed membranes after biofouling tests using copper and silver coated spacers [97] and, similarly, Ronen et al. used SEM images to study membrane fouling using anti-bacterial coated spacers [92]. Park et al., on the other hand, used SEM to visualize the membrane in a pilot study after a month long experiment [98]. Thomas et al. used SEM images to observe scalant deposition on MD membranes with different spacer designs [46,65] and Sreedhar et al. utilized SEM to view particulate fouling of the membrane before and after cleaning in the presence of spacers of different designs [64]. While versatility is an advantage of SEM imaging, it must be remembered that this is a highly granular tool and can give misleading information as the membrane area analyzed may not be representative of the actual state of fouling in the whole system. Therefore, it is difficult to draw significant conclusions merely using SEM imaging. In addition, SEM imaging requires a destructive sample preparation process, where the drying and coating of the membrane is necessary, thereby altering the state of the membrane and foulant layer. Nevertheless, it can be used as a supporting tool or for specific purposes such as corroborating a biocidal activity of spacer coatings by observing damages to the microbial cell [99].

2.2.2. Digital photography

It is often useful to take digital photos of the membrane, spacer, or

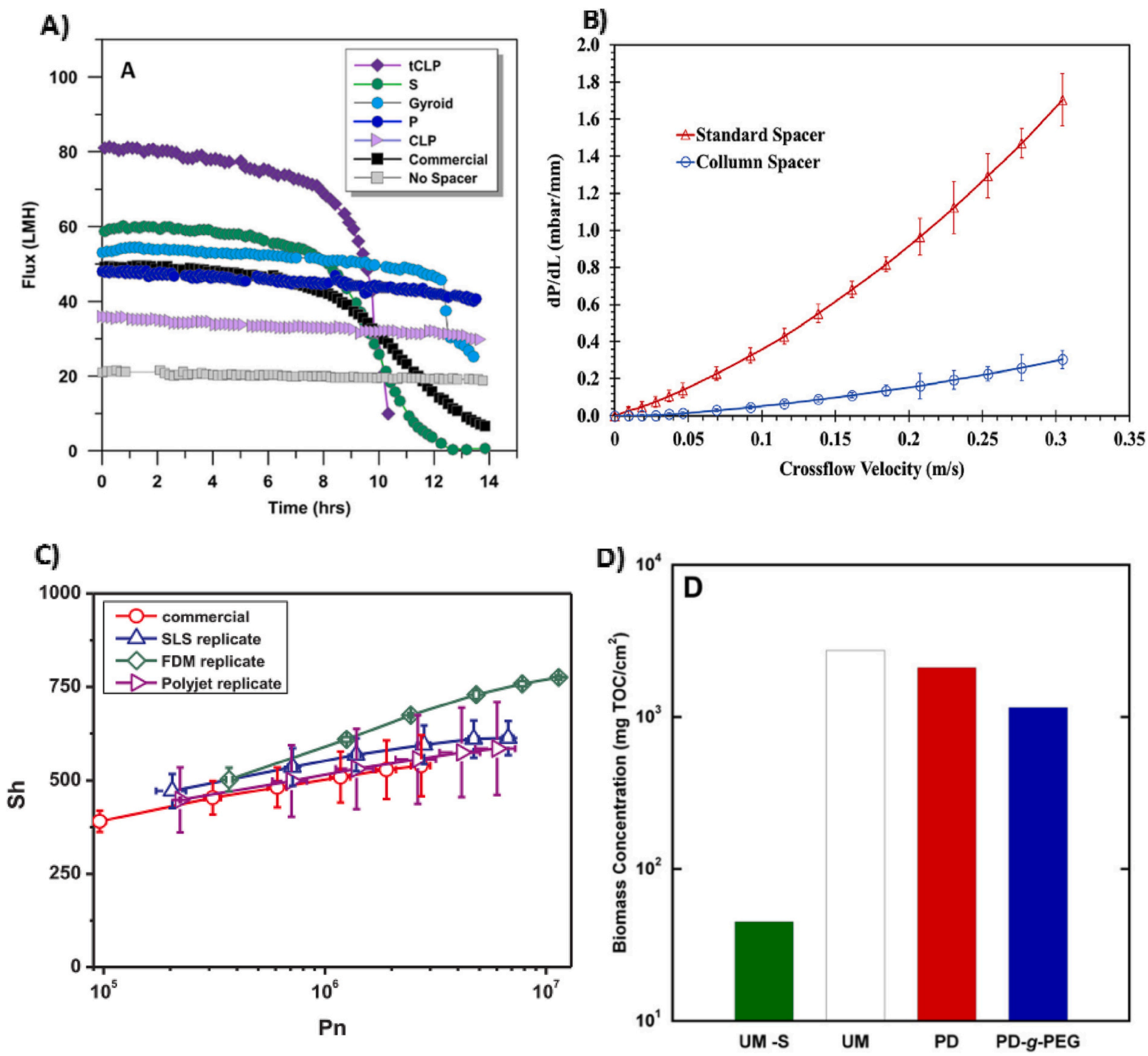

Fig. 7. Examples of different quantitative tools used for spacer evaluation: a) flux drop over time for TPMS spacers in MD, demonstrating the effectiveness of TPMS spacers [65]; b) feed channel pressure drop (FCPD) for standard and column-type spacers in UF filtration of seawater with sodium alginate and xanthan gum [73]; c) dimensionless numbers analysis of net-type spacers fabricated with various 3D printing techniques in UF of dextran solution [74]; d) total organic carbon (TOC) measurements after UF biofouling tests using uncoated membrane and spacer with no substrate dosage (UM-S), with substrate dosage (UM), polydopamine modified membrane and spacer (PD) and polydopamine-g-PEG modified membrane and spacer (PD-g-PEG) [42].

both to understand the effects of spacer on membrane fouling and one can find many creative examples of this n literature. Such images can be taken while the system is still in operation, which necessitate the use of transparent membrane cells. Alternatively, the images can be taken after the experiment. To name some examples, Linares et al. used photographs to show the variation of biofilm growth patterns using different spacer designs [100]. Massons-Gassol et al. captured photos of spacer and membrane coupons after fouling to show the biological growth on spacer filaments after operation [101]. Thomas et al. captured the intensity and patterns of calcium deposition on MD membranes using TPMS spacers by taking images of the scaled membranes after staining them with Alizarin red S dye, a compound traditionally used in histochemistry (Fig. 8(b)) [46,47]. Sreedhar et al. employed a similar strategy to detect biofouling on RO membranes after staining the membrane with crystal violet after tests with TPMS spacers. Their photos enabled an assessment of the impact various spacer designs had on biofouling [63].

2.2.3. Other tools (AFM, IR imaging, PIV imaging, etc.)

A variety of other visualization tools have been used in spacer research, which will be discussed here briefly. Particle imaging velocimetry (PIV) is a non-invasive tool that allows flow visualization in membrane cell to generate time-dependent velocity maps. This provides insight into the prevailing flow hydrodynamics in the cell in a way that can only be obtained otherwise through computational fluid dynamics (CFD). PIV has been utilized to visualize spacer-filled channels since the early 2000s [102,103] and was employed in the last few years to understand the flow in net-type spacer-filled channels [104,105]. It was also used in studying novel spacer designs. Haidari et al. used PIV to study zig-zag and cavity spacers and their flow in membrane channels [106] and to compare the ladder and diamond orientations of net-type spacers [107]. PIV was used in these studies to determine which property of the spacers, such as filament thickness or spacer height, had an effect on the hydraulic performance of the system. The variation of the fluid regime in the feed channel from steady to unsteady flow was determined, proving this tool to be useful in testing the accuracy of numerical simulations. Qamar et al. used PIV to understand flow

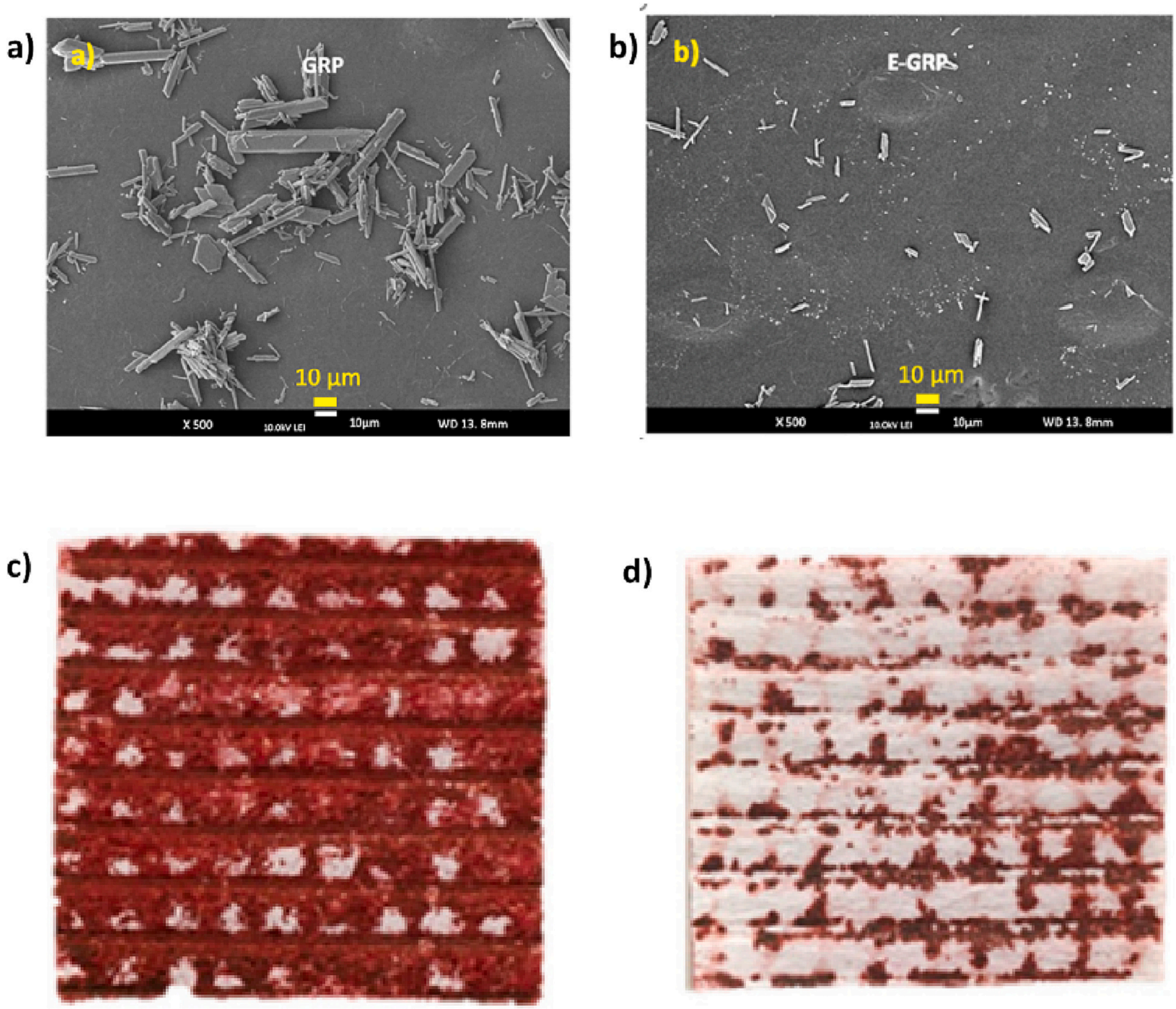

Fig. 8. Examples of two qualitative imaging tools used in spacer performance analysis: SEM images of commercial Toray FO membranes after inorganic salt filtration in the presence of a graphene-polylactic acid spacer with (b) and without (a) electrical polarization [49]. Digital photographs capturing calcium scale deposition on a PTFE MD membrane tested in the presence of an uncoated TPMS spacer (c) and fused-silica coated TPMS spacers (d), after staining the membrane with Alizarin red S dye [47].

regimes at different crossflow velocities in spacer filled channels [108].

Infrared (IF) imaging is a potentially useful tool in non-isothermal processes, such as MD. It was utilized by Anvari et al. to study the heating effect by externally heated stainless steel spacers in MD [48].

In 3D-printed spacers, the quantification of spacer surface roughness is important, as the latter plays a role in the attachment of biofilm and nucleation of scalants in the feed channel. Therefore, atomic force microscopy (AFM) and other surface roughness profilers are useful in studying these effects. Ronen et al. used AFM to study biofouling mitigation by zinc oxide coated feed spacers [109]. Tan et al. compared the roughness of different spacers printed with various 3D printing technologies and correlated it with the amount of biomass attached to the spacer [74]. Thomas et al. calculated micro-roughness, using AFM, and macro-roughness, using a 3D digital microscopy, for different 3D printed spacers with and without coatings, to capture their correlation with scalant deposition [47].

Most of the above mentioned tools require destructive ex situ analysis. A non-destructive in situ technique for fouling analysis is nuclear magnetic resonance (NMR). NMR offers an opportunity to study fouling in full-scale SWMs with a scan range of several mm^3, allowing both visualization and quantification of foulants including biofouling and organic fouling in spacer filled channels [30,110].

2.3. Combined tools

2.3.1. Fluorescence imaging

With the rapid growth in spacer-related biofouling studies since 2010, one of the most popular imaging tools has been fluorescence imaging. This can be 2D or 3D imaging, with the latter typically performed using confocal laser scanning microscopy (CLSM) [111,112]. The imaging is typically combined with other quantification processes such as cell counting or bio-volume determination, which can be conducted using image processing software such as ImageJ (Fig. 9(a,b)) [113]. To highlight some good examples, organo-selenium coated spacers developed by Vercellino et al. were analyzed using 3D CLSM imaging, and biomass accumulation was determined from the images

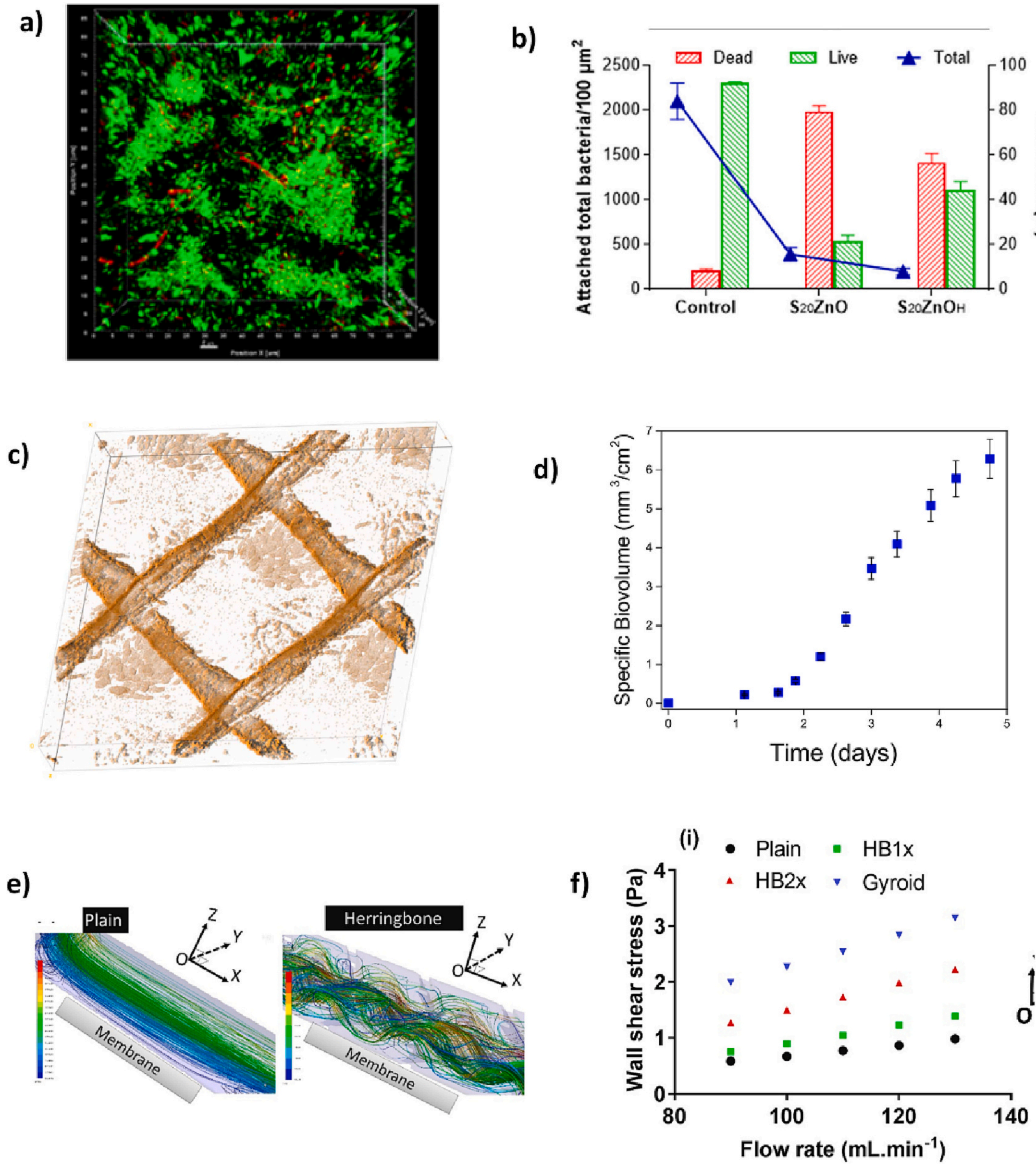

Fig. 9. Combined qualitative/quantitative tools used for spacer analysis: fluorescence imaging of membranes (a) combined with quantification of live/dead attached bacteria in the biofilm (b) [119]; c,d) OCT imaging of spacer and membrane (c) combined with biovolume measurements (d) [123]; and CFD analysis of flow in spacer-filled membrane channel (e) combined with shear stress calculations (f) [124].

[114]. Vercellino et al. also used 2D images in a similar study [115]. In 2013, Ronen et al. used CLSM imaging to study biofouling after using ZnO coated spacers in UF [109], and in 2015 the same group employed CLSM again, with biomass quantification, to study the biofouling mitigation capacity of nanosilver coated spacers [116]. Baek et al. used 3D CLSM images to study the anti-biofouling effect of electro-conductive spacers [117]. In 2018, Sreedhar et al. used a fluorescence microscope to take images of fouled membranes, followed by cell counting using the ImageJ software [63]. Yanar et al. printed spacers using different 3D printing techniques, and used CLSM images to study the build-up of the fouling layer in FO process under different scenarios [118]. Thamaraiselvan et al. coated polypropylene spacers with ZnO nanorods to induce anti-biofouling properties and used CLSM imaging and bacterial quantification to analyze its performance [119]. Kitano et al. developed carbon nanotube (CNT)-polypropylene feed spacers, at different CNTs concentrations, and tested them for organic fouling using fluorescence imaging. The fluorescence intensity was used for quantification of fouling activity [120]. Bogler et al. used fluorescent microscopy to study the initial deposition of fluorescent tracked bacteria and fluorescent beads in FO [121]. Recently, Thamaraiselvan et al. carried out a super hydrophobic coating on a polypropylene feed spacer using a poly (dimethyl siloxane) (PDMS) intermediate coating followed by candle soot nanoparticles, and employed CLSM imaging with biovolume determination for studying biofilm growth on the membrane [122].

Fluorescence imaging in general, and CLSM in particular, is a powerful tool for visualizing biofilms on membranes and spacers. However, this technique is limited by the necessity of staining with fluorescence dyes, which are always dependent on the amount and depth of staining. CLSM usage in membrane and spacer research has been mostly conducted as ex-situ analysis, by analyzing a membrane coupon after a filtration test [125,126]. This involves removing the membrane from the module followed by sample preparation, typically via staining with fluorescent dyes and membrane covering by cover slips. While this provides information about the biofilm thickness and coverage on the membrane, it also leads to loss of information on the sample during the preparation stage. In an interesting shift from the standard protocol, Tow et al. were recently able to successfully use CLSM to study membrane biofouling in-situ during MF and UF operations, which is a promising step towards scaling up CLSM application in membrane biofouling studies [127]. The new process still has certain limitations, such as the weakness of glass window that is required for viewing the membrane area, as well as optimized dosage of dyes, which can also affect the biofouling propensity. The limited sample viewing area was also noted, making it difficult to study biofouling over the full length of a module. There is also the added limitation that CLSM only allows the viewing of matter that can be stained by the appropriate fluorescent dye. However, these challenges can be overcome in the future to make CLSM a viable biofouling monitoring tool at large scale.

2.3.2. Optical coherence tomography (OCT)

OCT is a non-invasive tool of fouling visualization that has gained significant popularity in spacer research over the past decade. It can be used in-situ during process operation as long as a glass window is available for visualization, giving real-time insight into the development of the fouling layer (Fig. 9(c,d)). Several outstanding examples of such use of OCT are found in literature. In 2015, Wibisono et al. carried out one of the earliest OCT imaging on a spacer filled channel [128] followed with another study using OCT imaging of a hydrogel modified feed spacer to minimize fouling [70]. In 2016, West et al. carried out 3D OCT imaging to calculate the biomass on RO membranes with spacers [129]. In 2017, Fortunato et al. were able to determine the bio-volume deposit on membranes from OCT images, thus confirming the impact of spacers on biofouling [123]. They were also able to map the biofilm thickness later that year [130]. In recent years, several studies that developed novel 3D printed spacers utilized OCT images in their analysis of spacer-induced fouling mitigation. The studied spacer designs include perforated [67], column-type [73,131], helical filament [68], dynamic turbospacers [69,88], honeycomb shaped [87] and hole-pillar [132] feed spacers.

While OCT imaging has been mainly conducted for biofouling analysis, its application is expanding. For example, in 2021, Wong et al. employed OCT in situ in an MD system using textile wastewater as feed to evaluate the growth of fouling layer thickness for a mixture of dyes [133]. OCT has also been employed in the evaluation of scaling in MD [134]. Of particular benefit to MD, OCT was employed to visually locate areas of wetting in MD membranes by identifying areas where scale formation has occurred inside the pores [135]. While the scan area of OCT is still a limiting factor, the advantages of this technique in terms of the range of applications that can be tested, the different forms of fouling that can be measured, the ability to do time-series quantifications, and its combination of qualitative and quantitative analysis, all make OCT an excellent candidate for studying spacers. The adaption of OCT imaging for spacer research in membrane science is expected to grow in coming years.

2.3.3. Computational fluid dynamics (CFD)

CFD modeling is another powerful tool that allows the combination of qualitative and quantitative analyses of spacer performance. While details of CFD modeling protocols for spacer filled channels lies outside the scope of this review, it is nonetheless worth mentioning some notable examples where CFD has been combined with experimental analysis to study novel spacer designs (Fig. 9(e,f)). CFD modeling of flow in feed channels with sinusoidal spacers was carried out by Xie et al. in 2014 [136]. In 2014, Radu et al. studied particle deposition in a spacer filled channel experimentally, and corroborated their results using CFD simulations [39]. Ronen et al. carried out numerical simulations to study the release of silver ions from coated zig-zag spacers [116]. CFD analysis was also carried out for microspacers in microfluidic channels, as well as for vibrating spacers [124,137]. Novel spacer designs such as perforated spacer [67], helically symmetrical spacer [68], and modifications of net-type spacer (such as those performed by Siddiqui et al. [93]), were carried out using direct numerical simulation (DNS) analysis as well.

CFD analyses can help bridge the gap between the design and testing phases of novel spacers' development [138]. They can also be used to determine the values of dimensionless numbers such as Sherwood number and Power number related to spacer performance, which can be then used to determine parameters such as the mass transfer coefficient, flux, rejection, and pressure drop in large-scale systems [139]. CFD models were also developed to study various fouling phenomena in membrane modules, including biofouling and organic fouling [37,140].

2.4. The scale issue

When selecting a spacer characterization technique from the wide range of tools available for this purpose, it is necessary to keep in mind the scale at which the observations are made, to avoid misleading results. As can be seen in Fig. 10(a,b), different tools can vary significantly in their scale of observation. On one end of the scale spectrum is SEM imaging (Fig. 10(c)), which allows the highest magnification and the ability to examine the membrane's cross-section. The caveat is that in the same membrane sample, it is possible to obtain SEM images of regions with absolutely no microbial growth at the microscale (μm), but upon examining photographic images of the membrane, the latter may reveal a surface heavily covered with bacterial colonies in many regions, albeit outside the SEM's field of view.

Like SEM, CLSM suffers from a similar problem of high resolution but low sample area (Fig. 10(d)). On the other end of the spectrum, a holistic, macro-scale view (on the order of few to tens of centimeters), such as that obtained from a digital photograph or CFD modeling, may provide insight into fouling patterns, but will be insufficient to understand how the foulants' deposition and attachment take place at the microscale (Fig. 10(e)). As a meso-scale tool (on the order of few millimeters), OCT is highly helpful in this regard, particularly because it also has a 3D component which allows the examination of foulant layer in the z-scale (Fig. 10(f)). It is worth noting, however, that OCT requires an open field of view, which renders it unusable in the case of spacers with highly dense structures, where the membrane surface may not be visible. OCT also has a limited scan area and the analysis of membrane fouling beyond a few spacer filaments cannot be carried out. As such, it may not be possible to use OCT to study the development of fouling along the length of the full module.

3. Spacer research since 2010

Early spacer research was largely focused on identifying the appropriate means to characterize spacer performance [141], as well as identifying the effect spacers have on parameters such as flux and critical flux [22], mostly via CFD simulations [79,142–145]. In contrast, spacer research since 2010 has focused greatly on spacer modification (both geometrical and surface modification), with the majority of these reported modifications aimed at fouling mitigation. While novel spacer geometries have been tested during the early stage as well, the chemical modification of spacer surfaces was a new trend seen only since 2010, and until recently it was aimed solely at biofouling mitigation. Fig. 11(a, b) shows a distribution of research publications from 2010 to 2021 within two main categories of spacer modification studies; chemical and geometry modifications.

3.1. Chemical modification of spacers

The coating of membranes for biofouling mitigation has been a long-standing research approach. The critical step in forming a biofilm is the initial attachment of the microbes onto the surface. Therefore, the propensity of microbial attachment can be minimized by altering the membrane's surface properties such as charge, roughness or hydrophilicity. Another strategy is to induce anti-bacterial properties, which can

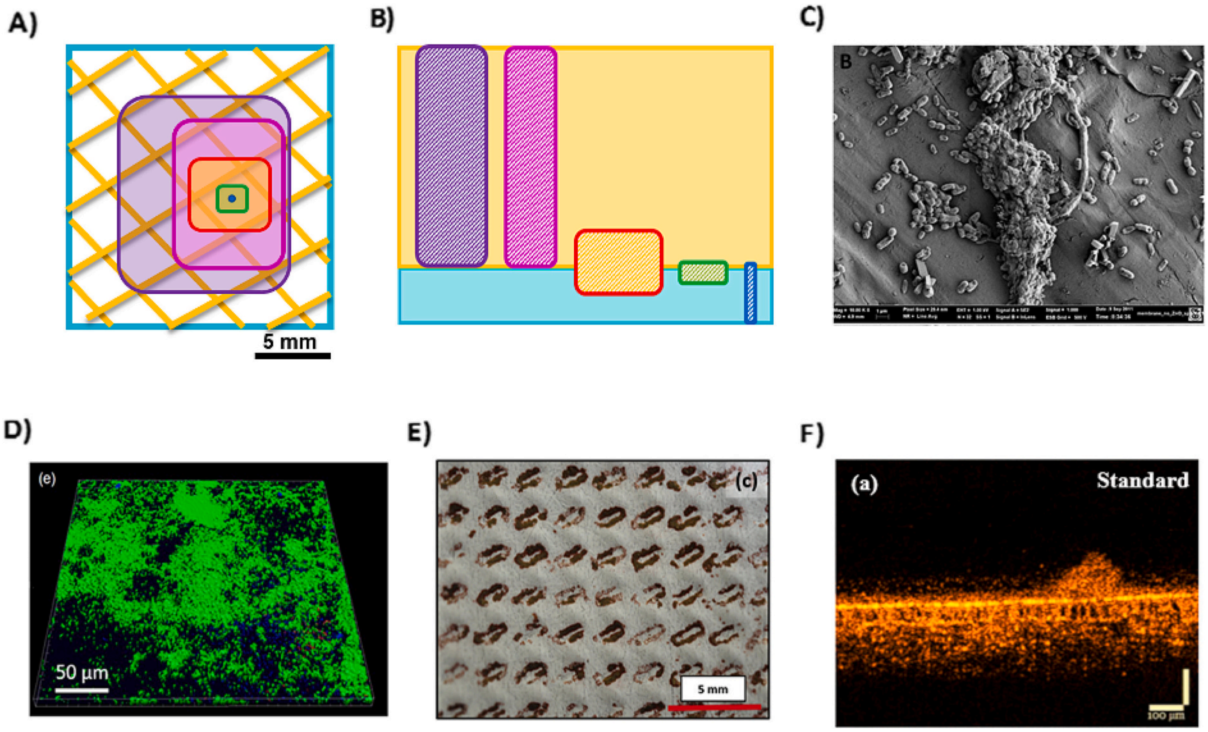

Fig. 10. The issue of scale in visualization tools used in spacer research: a,b) top-view and side-view schematics images depicting the relative scale of five visualization tools (purple: CFD analysis; pink: digital photography; yellow: OCT; green: CLSM; blue: SEM); c) SEM image of UF polysulfone membrane after biofouling tests with an unmodified spacer [109]; d) CLSM image of fluorescent dye stained poly(dimethylsiloxane) coated spacer after biofouling tests [122]; e) digital photo of alizarn red S stained PTFE membrane after MD scaling tests when using a gyroid TPMS spacer [46]; f) OCT 2D scan of foulant deposition on PES membrane after UF filtration tests with a standard diamond spacer [68].

act as a fail-safe in the case of bacterial attachment [29,146]. Given that earlier mentioned consensus that feed spacers are often the point of initial microbial attachment, it stands to reason that modifying the spacer properties can be an advantageous route to reduce biofouling.

The materials reported in the literature for antibacterial surface modification of spacers could be broadly classified as biocidal heavy metals and their derivatives or organic and polymeric materials. The biocidal properties of heavy metals is caused by several metal-catalyzed oxidation reactions that damage microbial proteins, membranes, or deoxyribonucleic acid (DNA) [147]. Anti-bacterial and anti-adhesion properties of copper coating on polypropylene feed spacers was reported to lower the development of extracellular polymeric substances (EPS) on the membrane but at the expense of reduced flux [148]. The deposition of zinc oxide (ZnO) on the feed spacer is noted to lead to the microbial death on the membranes [109]. Silver is by far the most commonly used antibacterial metal/nanomaterial, but it is extremely susceptible to water leaching, which is unavoidable in membrane applications [149]. Although silver nanoparticle coating on feed spacers reduces biofouling prior to leaching, it is reported that leached silver nanoparticles eventually cause colloidal fouling on the membrane [97]. However, the leaching of sonochemically deposited silver nanoparticles from the feed spacer has also been reported to adversely affect the growth of bacteria on the membrane and within the feed channel [116]. Other aspects investigated regarding surface modification of spacers by metallic coating include the effect of morphology and the combined use with hydrogen peroxide. The use of ZnO nanorods coated feed spacer, as opposed to the amorphous ZnO coatings, reduced biofouling by inducing mechanical damage to the microbes [119]. Yang et al. utilized copper oxide (CuO) coatings on feed spacers, and while these were also expected to have an additional effect through Fenton reaction with hydrogen peroxide (added for bubbling effect for cleaning), the results were attributed to the dosage of hydrogen peroxide alone, with the coating proving ineffective [150].

Non-metallic (mostly organic or polymeric based) coatings typically exploit the hydrophilic properties of these compounds to achieve antibacterial activity. Coating of spacers with non-metallic elements, such as organo-selenium, can lower biofouling but at the expense of reduced flux [114,115]. The modification of polypropylene spacers by a hydrophilic agent such as diglyme resulted in positive antibacterial results [151]. The coating of feed spacer with three-dimensional (3D) network of hydrophilic polymers, known as hydrogels, with varying surface charge (neutral, cationic, anionic) demonstrated that anionic hydrogel (polySPMA) showed the best results in reducing biofouling, which could be due to its high hydrophilicity [70]. Similarly, a feed spacer coating with a well-known superhydrophilic biofouling material, zwitterionic poly(sulfobetaine methacrylate), showed successful short term batch results in preventing bacterial attachment, but long term results showed no significant effects on the coated spacer [72]. Organic fouling is reported to be minimized by using CNT-polypropylene nanocomposites in making net-type spacers [120]. Contrary to the popular belief of using hydrophilic materials to repel organic materials and bacterial attachments, use of superhydrophobic polymeric substances such as an intermediate PDMS coating followed by candle soot nanoparticles was also shown to minimize biofouling by trapping air microbubbles within the coated surface [122].

Few studies have compared the performance of spacers with metallic and organic/polymeric surface modifications. Spacer coating with silver nanoparticles exhibited a long-distance antibacterial activity when compared to the localized antibacterial activity displayed by grafting polymeric quaternary ammonium [92]. In a study conducted by Rice et al. in 2018, silver nanoparticle coating demonstrated the best antibacterial activity when modifying polypropylene spacers for RO in comparison to other coatings like silver, silica nanoparticles, and graphene oxide, which covered a range of properties like biocidal, anti-adhesive, hydrophilic, etc. [152]. It is worth noting that in this study, Rice et al. also determined the surface energy and contact angle of the

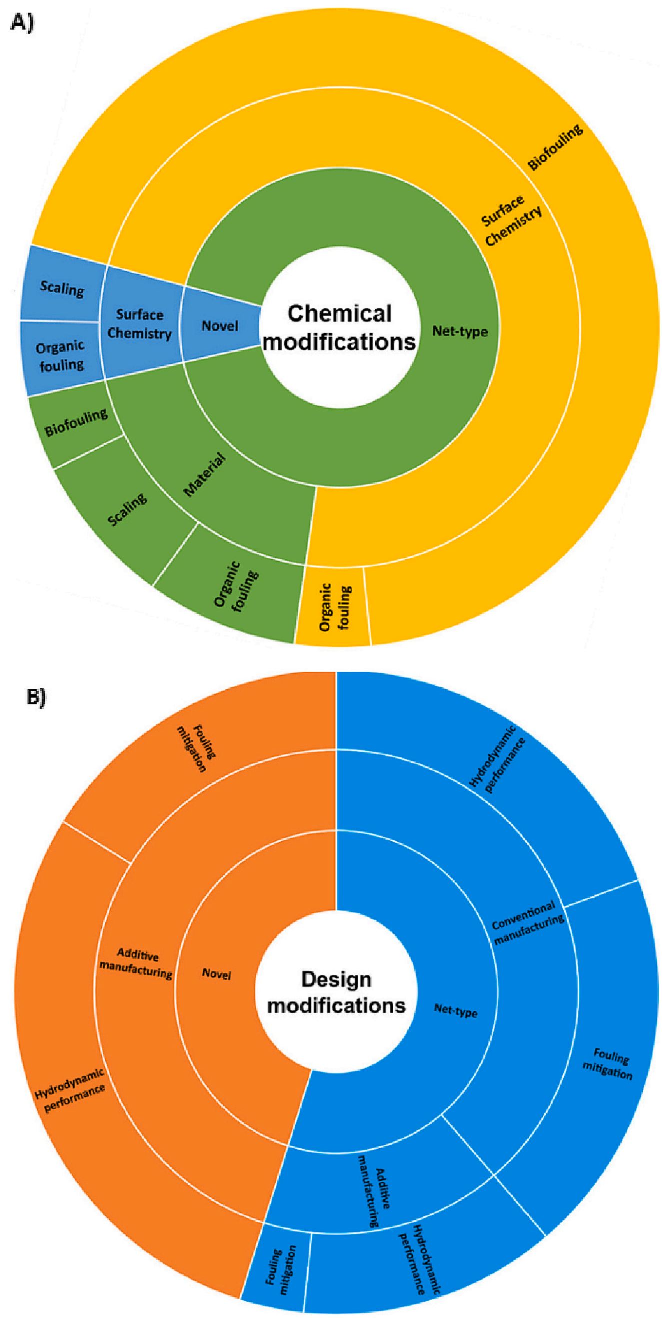

Fig. 11. Breakdown of research publications (by fraction) within the two main categories of spacer modification studies from 2010 to 2021; a) publications reporting on chemical modifications of spacers (total: 26 papers); b) publications reporting on geometry-based modifications of spacers (total: 31 papers). Distribution is shown for several categories and subcategories for each modification approach.

spacers and incorporated these properties in their analysis, and these characterizations are generally missing from modified spacer studies. It is also important to establish the effectiveness of antibacterial activity of surface modified spacers based on long term studies. Araujo et al. compared the effects of coating polydopamine, polydopamine-*graft*-poly (ethylene glycol), copper coating and commercially available biostatic (triclosan) on polypropylene feed spacers, which is a combination of hydrophilic and anti-bacterial materials. The study showed that none of the coatings were effective in addressing the problem of biofouling in the long-term [42].

In general, coating of feed spacers produces mixed results that are typically positive; however, there are additional concerns that must be addressed, such as new manufacturing procedures, leaching, and the use of expensive or environmentally damaging materials. In the coming years, there may be greater emphasis on 3D printing of blended materials, which not only reduces the number of steps needed to make the spacer but also produces a more robust spacer with less leaching than surface deposition. Something interesting to note from Fig. 11a is the dire lack of chemically modified spacers with novel geometries. The combination of novel geometries with antifouling chemistry is an interesting avenue to explore. Certain coatings that pertain to unique applications will be discussed in Section 4.2.

3.2. Spacer modification based on geometry

The modification of spacer geometry offers a passive method to improve hydrodynamics and minimize fouling in the membrane channel. Novel spacer geometries have been proposed for a while, even though the majority have been based on CFD modeling. Nevertheless, there were some novel spacers prepared even prior to the 3D printing boom seen since 2013. Currently, 3D printing technologies are increasingly capable of replicating complex spacer designs at the required scale and are constantly improving [14].

In 2004, Li et al. carried out one of the earliest studies on novel spacer geometries by developing innovative spacers with modified filaments, twisted tapes and multilayer structures, which were tested experimentally [10]. Mass transfer analysis was performed using the limiting current method, revealing that the multilayer spacer demonstrated 30 % higher Sherwood number and 60 % lower power consumption than the commercial feed spacer. By 2008, Shrivastava et al. developed herringbone, helical, and ladder-type spacers using fused deposition modeling (FDM) to target CP and provided a framework to optimize spacer design [153]. These spacers were also analyzed using the electrochemical method of mass transfer analysis, which may not be well-suited for RO and UF [14]. Fritzmann et al. developed novel double helical micro-structured feed spacers printed using Polyjet technology based on the Kenics static mixers. Their spacers showed a doubling of critical flux in tests conducted in air-sparged membrane systems [62]. They further experimented with these spacers in UF and studied the effect they could have on membrane selectivity [154]. In 2016, Siddiqui et al. carried out 3D printing of net-type spacers that showed reduced biofouling and improved hydrodynamics [93]. This study not only demonstrated the feasibility of using 3D printing for spacer development by corroborating numerical simulation with experimental results for a given spacer design, but also confirmed the accuracy of 3D printing in replicating a commercially fabricated spacer design. They also modified the net-type design, which was 3D printed, and the modified spacer showed enhanced biofouling mitigation. Li et al. proposed a novel hairy spacer with vibrating fibers that was tested in FO, showing enhanced mass transfer [155]. In 2017, Tan et al. conducted studies on 3D printing of spacers with various commercial 3D printing technologies which helped advance the understanding of spacer fabrication with different processes [74,156]. The commercial spacer was compared to 3D printed spacers made from selective laser sintering (SLS), FDM and Polyjet technologies using mass transfer analysis by dextran filtration. The Polyjet spacer showed highest replicability of the CAD design, while the FDM spacer had superior results due to irregularities introduced into the spacer during the printing process. They also demonstrated the effect of the different materials and their roughness on bacterial attachment. Other unique feed spacer designs that have been investigated include sinusoidal and saddle shaped spacers [157], and vibrating spacers with novel geometries (Fig. 12(a,b)) [137,158].

Feed spacers were developed using the triply periodic minimal surface (TPMS) architectures and were tested in a range of operations such as UF and RO for mass transfer enhancement (Fig. 12(c,d)) [63,65]. In both UF and RO, the Gyroid TPMS design improved flux and decreased biofouling in comparison to the traditional net-type spacer. During mass

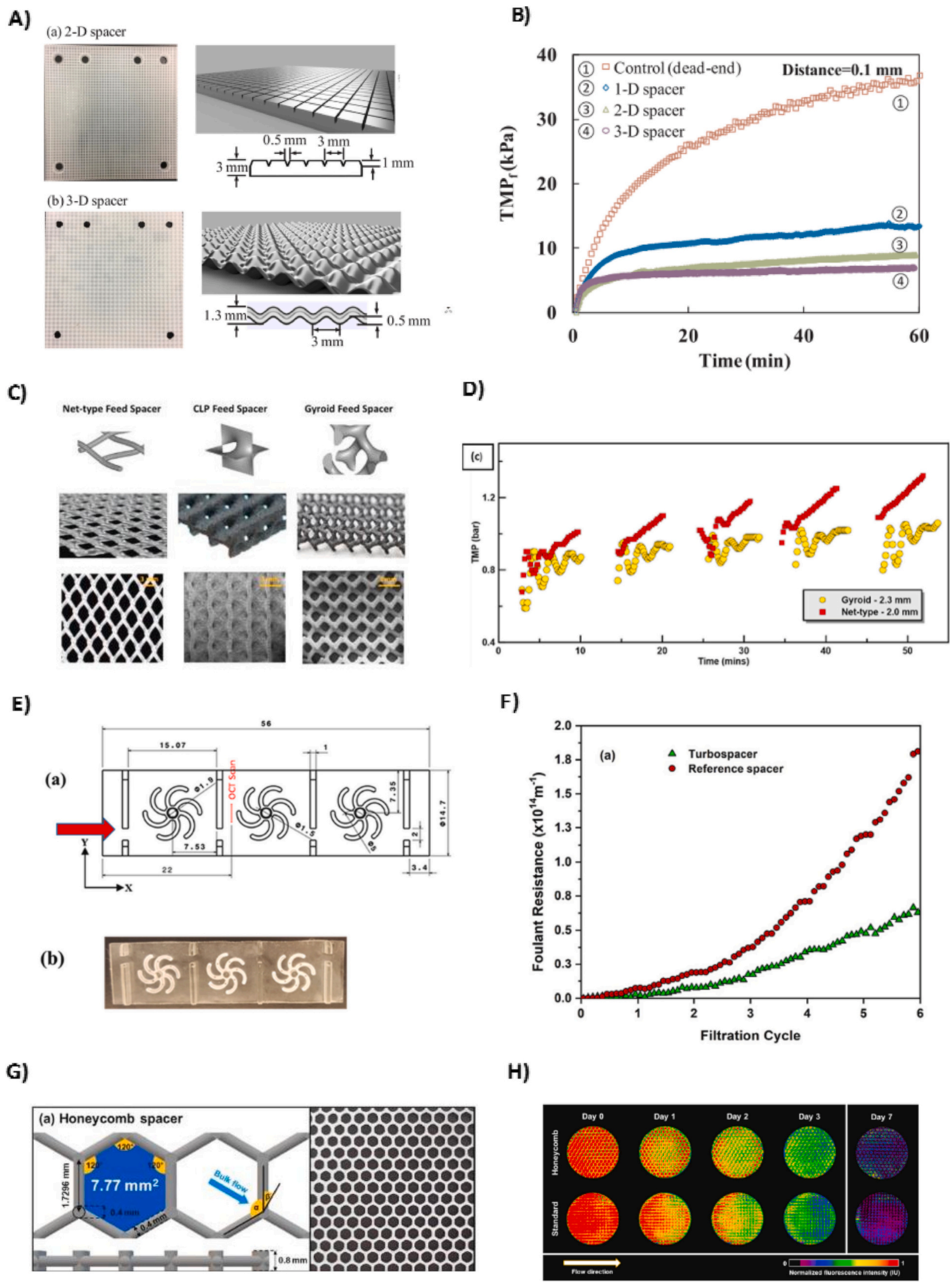

Fig. 12. Novel spacer geometries tested using various performance evaluation techniques: a,b) vibrating spacers tested in UF and evaluated via TMP change with time [137,158]; c,d) TPMS spacers evaluated in UF via membrane cleaning efficiency over 5 cleaning cycles [63,65]; e,f) dynamic turbospacers evaluated in FO via fouling resistance using a TFC membrane and primary wastewater effluent as feed [69]; g,h) honeycomb spacer evaluated in NF via fluorescent imaging of fouling deposition after humic acid filtration [87].

transfer studies, the TPMS spacers showed enhanced mass transfer, though they also encountered increased energy losses. The Gyroid and Crossed Layers of Parallel (CLP) designs were promising and selected for further studies. The TPMS spacers were also tested for heat and mass transfer enhancement and scaling and organic fouling mitigation in MD [46,66]. In MD, TPMS spacers had 60 % higher water flux and 63 % higher heat transfer coefficient than the net-type design. During scaling mitigation tests using calcium sulphate, the transversely arranged CLP (t-CLP) showed 50 % higher flux than the net-type spacer, but at the expense of increased pressure drop. A promising hybrid TPMS design of both t-CLP and Gyroid architecture was then tested and showed scaling mitigation similar to the tCLP but with lower pressure drop. In 2019, Castillo et al. also tested the TPMS spacers for organic fouling in MD and reported reduced organic fouling and 30-70 % higher water flux using the Gyroid and t-CLP, compared to net-type spacer.

The TPMS spacers were further tested for their impact on backwash and cleaning efficiency in UF [64]. Sodium alginate filtration was carried out for multiple cycles, interspersed with backwash/relaxation procedures, and a rise in TMP over 20 cycles was observed. The feed spacers were shown to have a significant effect over the change in pressure, with the t-CLP spacer showing minimal cake formation and enhanced backwash efficiency. These spacers were further developed by surface modifications to induce antiscaling properties in MD [47] and photocatalytic cleaning properties in UF [159]. Thomas et al. also compared the TPMS spacer to commercial net-type spacer for the air-gap MD process, however the mass transfer enhancement was reduced when compared to the results for the direct contact MD experiments [8]. The mass transfer resistance due to the air gap is believed to have reduced the influence of the spacer in AGMD system.

A range of other novel spacer geometries developed in recent years are briefly herein described. In 2018, Kerdi et al. [71] tested perforated feed spacers, which feature net-type designs with 1- to 3-holes. The 1-hole perforated spacer demonstrated the greatest flux improvement while the 3-hole spacer had the least pressure drop when compared with symmetric spacers without perforations. The same perforated spacer design was also tested in FO but its resultant improvement was less significant compared to UF due to the nature of the driving force of the process [160]. In 2019, Ali et al. developed column type feed spacers with spacer nodes shaped like columns, which demonstrated a 66 % lower pressure drop than a regular spacer for UF applications [73]. In 2021, another spacer with combined features of the column-type, perforated, and hole-pillar spacer designs was developed by Qamar et al. [132]. The hole-pillar spacer showed up to 75 % reduced pressure drop during low pressure filtration, as well as minimal biofilm development during the biofouling tests. In 2020, Kerdi et al. also developed helical filament spacers where the degree of twisting (number of helices per filament being 1, 2 and 3 helix) was controlled and compared with a spacer with no helices in UF [68]. The 3-helical spacer had the highest flux with up to 65 % lower pressure drop in UF tests, while also demonstrating biofouling mitigation. In 2020, Ali et al. designed a novel dynamic turbospacer, which has a series of turbine-like elements in a ladder-type configuration that enhanced flow and increased unsteady flow in UF (Fig. 12(e,f)) [69]. The spacer showed promising results both in fouling mitigation and hydraulic performance, with significantly reduced pressure drop and 300 % higher specific flux during UF tests with sodium alginate, xanthan gum and microbial seawater. In 2021, the dynamic turbospacer was subsequently tested in an FO process with wastewater plant effluent as feed and exhibited promising results in that application as well, with much lower cake resistance [88].

In 2021, Park et al. developed a novel spacer for NF application, with spacer filaments shaped like a honeycomb (Fig. 12(g,h)) [87]. The spacer showed increased permeate flux as well as reduced fouling layer, as visualized using OCT, which was also confirmed by the lower cake resistance. In 2021, Dang et al. printed spacers with complex geometries at microscale for microfluidic filtration, with previously used designs such as the herringbone and TPMS [124]. The Gyroid based design showed significant flux enhancement of up to 93 %, with a 23 % higher pressure drop as trade off.

4. Future outlook

3D printing will continue to play a role in shaping spacer research in the near future. A new application for 3D printing in membrane research is to create patterns on the membrane surface [161] for increased localized unsteady flow and fouling mitigation. Membrane patterns, optimized using CFD modeling, can be printed indirectly with the use of a printed mould and result in increased flux and reduced organic fouling [162,163]. These patterns play a similar role to feed spacers, and are an integrated unit that blurs the distinction between the membranes and the spacer. One aspect to take into consideration is that research on spacer surface modification tends to lag behind membrane chemical modifications and coatings. In this section, three aspects of the future outlook for feed spacers are discussed.

4.1. Scale-up of novel spacers

Despite the interest in developing novel spacer designs, research on this topic has been criticized for failing to address the practical aspects of using these spacers in SWMs, especially the possible damage to the membrane during rolling into an SWM module or during high pressure operation [164] and the lack of agility of some of these designs during the said rolling. This is particularly relevant for 3D printed spacers with complex geometries. Most novel spacer studies completely fail to address the practical challenges of using these designs in membrane modules. Spacers and membrane leaves undergo significant stresses during the module rolling and subsequent operation of these SWMs. Indeed, the thickness of the spacer is reduced by the compressive forces generated during the roll up [2]. It is also important to remember the primary function of the spacers, which is to adequately separate the membrane leaves. The density of the spacer support points is closely linked with the thickness of the channel, and a spacer with reduced points of contact could lead to a very compressed channel. Hence the thickness of the spacer, its density and its points of contact are all important features. While currently these parameters such as density have been considered in terms of spacer performance, they must also be considered based on potential scale-up. On the other hand, the forces employed in compressing the module can also lead to damage of the membrane surface by the sharp edges in certain spacer designs, or else deformation of the spacer itself.

The most pertinent question remains the scalability of these designs to an industrial production scale. To the best of the author's knowledge, to this date, none of the novel spacer designs fabricated and tested at lab scale have been fabricated at an industrial scale and implemented into a commercial SWM. Fig. 13 shows a summary of various novel spacer geometries printed, and the range in which they have been fabricated and tested.

As described in Section 3.2, spacer geometry modifications broadly fall into two categories, spacers that are modified net-type or similar to net-type design (such as honeycomb spacer, perforated spacers etc.), and spacers that have significantly different and complex architectures (TPMS spacers, turbospacers etc.). In the first case, 3D printing can be used to test spacer prototypes at lab-scale but proven production processes, such as extrusion, can be used for industrial-scale manufacturing. This can be considered a major advantage for these spacer designs, due to their ease in scale-up, as it follows the current conventional wisdom of using 3D printing for prototyping before advancing to conventional manufacturing processes for mass production and economics of scale. This strategy was promoted by Siddiqui et al. in their study on modified net-type designs [93], although it may not necessarily be limited to these designs. Novel designs that resemble the net-type spacer, such as the column-type [73] and honeycomb [87] spacers, can potentially be fabricated using existing manufacturing processes, which can make their

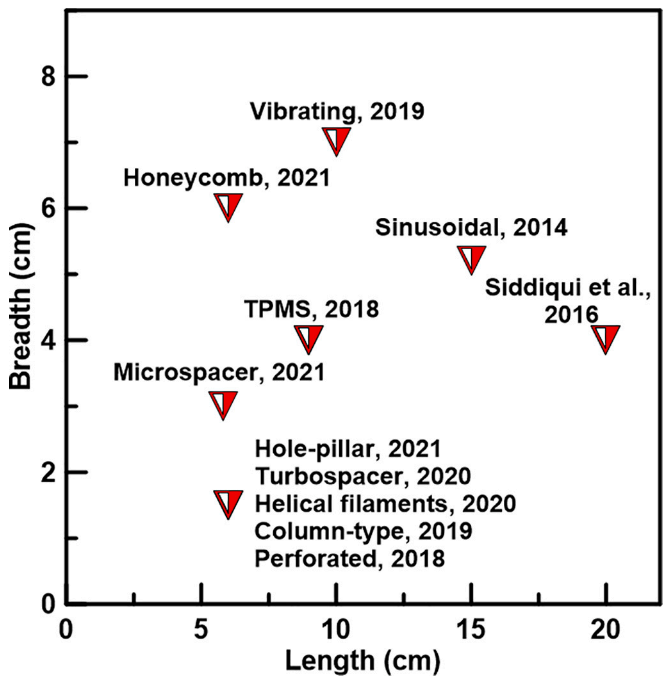

Fig. 13. Surface dimensions (breadth versus length) of notable novel 3D printed spacers reported in literature from 2010 to 2021 (Note: some were obtained based on the reported membrane area).

adoption at a large scale more feasible. Indeed, if novel spacers are to be adapted by the industry, this pathway remains the most likely to achieve that result.

On the other hand, the scalability of complex novel designs, such as TPMS spacers and turbospacers, to industrial scales is heavily dependent on the advancement of 3D printing technologies to overcome the current limitations pertaining to printing scale, speed and cost. Fig. 13 depicts that the size of most novel spacers that have been fabricated for lab-scale testing is significantly smaller than the size required for commercial spacers. With regards to speed, the conventional extrusion process can fabricate materials at speeds of around 640 m/h, while industrial 3D printers fall short significantly, at around 0.05 m/h [18]. Indeed, with increased design complexity, a greater stress is placed on the printer speeds and the computational power required for the CAD designs. The cost of 3D printing is another significant bone of contention, given that commercial spacer costs are extremely low (below $1.00/m^2) [2] and can be prohibitive factor unless there is a significant decrease in 3D printing costs. For a process such as FDM, the cost of plastics used can be as high as US$ 250/kg, which does not take into account the high cost of large-scale printers [165]. Although these costs have fallen exponentially over the past few decades and are expected to continue to do so in the coming years, 3D printing spacers on an industrial scale will still require significant improvements in the layer-by-layer printing speeds of 3D printers as well as more effective polymer use. Some of these costs may be mitigated by the potential benefits of the innovative spacers in the long-term operation of the plant, including increased permeate production, extended membrane life, and lower cleaning expenses. Lastly, the role and selection of 3D printed spacer material needs to be considered as well. As stated earlier, any material used needs to be flexible enough to fold into a SWM tightly. For pressure driven processes, the mechanical strength of the material is a valuable property, while for heat-driven processes the thermal stability of the material will be significant. The toxicity of the material must be considered for water treatment and desalination applications as well. These challenges will be met as greater variety of materials emerge as options for 3D printing technologies.

4.2. Rise of functionalized spacers

The utilization of feed spacers for additional functions beyond the traditional role of supporting the membrane and enhancing unsteady flow has been attempted, largely through passive chemical coatings aimed at reducing biofouling, as discussed in the previous section. Fig. 14(a) illustrates the growth in spacer functionalization studies, in comparison to the other two forms of spacer modification (geometry and chemical). In functionalized spacers, the coating is expected to perform a role other than passively minimizing membrane fouling. An early example of functional spacers was presented by Baek et al. in 2014, where stainless steel meshes were used as electrically conductive feed spacers that could be periodically polarized to cause the detachment of bacteria from the spacer and membrane surface [117]. In 2017, Abid et al. used electrically conductive spacers to carry out electrolysis and generate microbubbles for fouling reduction in membrane systems [166]. Anvari et al. designed stainless steel spacers that could be heated through induction by radio frequency, which enhanced flux and reduced TP in MD (Fig. 14(b)) [48]. In 2019, polypropylene spacers functionalized with copper oxide coatings were reported, with the intention of generating radicals in the presence of hydrogen peroxide for the targeted removal of biofouling on the membrane (Fig. 14(c)). A similar approach was reported by Kerdi et al., where biofilm was successfully removed from the spacer and membrane by using direct electric current that generated a controlled amount of chlorine [167]. In 2021, Yanar et al. prepared feed spacers by 3D printing using a blended polymer [49]. Polylactic acid (PLA) was blended with graphene and 3D printed into net-type spacers, and subsequently polarized in an electrical field. These spacers showed great antiscaling properties in FO operation with gypsum scaling, and is an early example of using 3D printing to incorporate modified chemistry into the polymer itself, which can offer several advantages [49]. The charged spacers attracted the scalants onto themselves instead of the membrane. In 2021, Thomas et al. coated feed spacers with fluorinated silica nanoparticles through the sol gel process in order to reduce scaling on both the spacer and membrane during MD operation (Fig. 14(d)) [47]. The spacers were tested using calcium sulphate foulant in direct contact MD system and showed 70 % and 60 % reduction in scaling on the spacer and membrane, respectively. This was attributed to the change in the spacer's surface free energy after modification, which affected the scale formation in the feed channel. In 2022, Sreedhar et al. developed a feed spacer coated with β-FeOOH nanorods that has photocatalytic activity for decomposition of dissolved organics in the feed stream and foulant removal off the UF membrane (Fig. 14(e)) [159]. The t-CLP spacer was coated with β-FeOOH nanorods and showed significant decomposition of 4-nitrophenol and methylene blue under solar light illumination.

4.3. 4D printing and smart materials

The field of additive manufacturing is currently undergoing a rapid rise in interest in smart materials and 4D printing, which is already garnering interest in various fields such as healthcare and aerospace [168]. It would be reasonable to expect this new field to influence membrane and spacer research in the coming years as well.

4D printing refers to the development of materials which can respond to an external physical or chemical stimulus, such as electricity, light, temperature, etc., which is then printed into the desired architecture. These can involve various materials, including shape memory polymers (SMP), shape memory alloys (SMAs), and shape memory composites (SMCs) (some examples are shown in Fig. 15) [169].

4D printed materials that are attuned to the desired stimulus can potentially provide a range of functions in the spacer channel, and they can be broadly classified into two categories, namely shape shifting and shape memory materials. Shape shifting prototypes change their shape when an external stimulus is applied, and immediately return to their original shape when the stimuli are removed. Shape memory prototypes

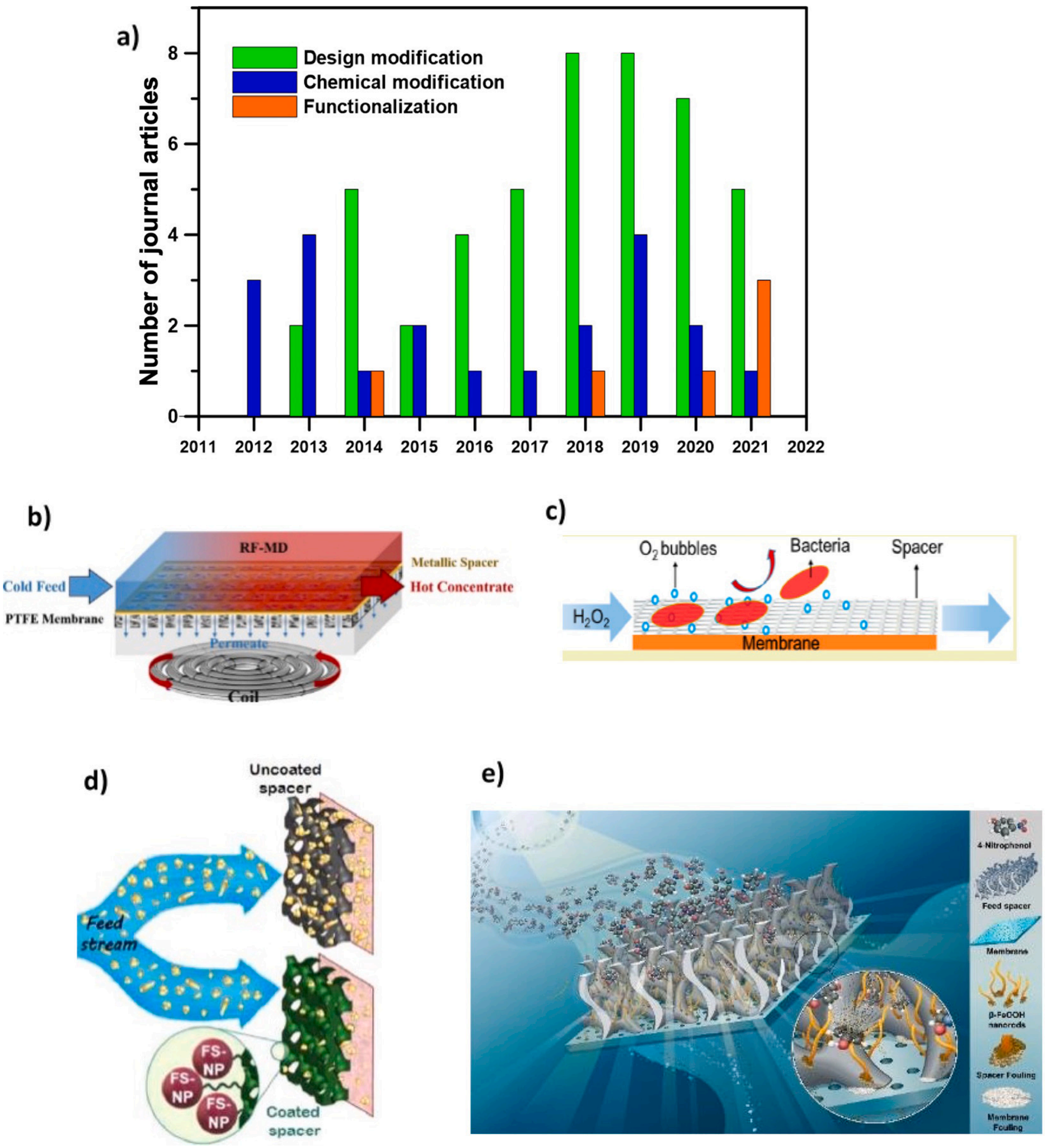

Fig. 14. Growing trend of functionalized spacers in water research; a) increased functionalized spacers in research over the period of 2010 to 2021; b) stainless steel spacers heated through induction by radio frequency for MD [48]; c) CuO coated spacer capable of fenton reaction cleaning for biofouling mitigation in the presence of hydrogen peroxide [150]; d) feed spacers coated with fluorinated silica nanoparticles inducing antiscaling properties in MD [47]; e) photocatalytic feed spacer for decomposition of organic pollutants in the feed and organic fouling mitigation in UF [159].

can hold their shape until a different stimulus is provided to return to the original shape. These are collectively known as smart materials, and the selection of the right materials can lead to a range of applications [18]. While 4D printing is yet to be applied for spacers, there are some potential applications such as spacers that respond to certain lapsed time or increased pressure to change shapes and can potentially increase the lifetime of the module by opening new flow channels for the feed after significant fouling. Optically activated spacers can be used to sporadically clean the feed channel through movement in a manner that mimics photocatalytic cleaning without the associated issues such as catalyst degeneration and membrane aging with radicals formation. Time dependent smart spacers can be designed to extend the lifetime of the membrane module by timing the opening within the spacer geometry to increase the size of the flow channels after a certain extent of fouling has occurred. Electrical or magnetically influenced materials can also be designed to induce shaking or vibrations in the feed channel on introduction of an electric current, leading to membrane cleaning. pH sensitive materials can be used in spacers that are activated when biofouling or other types of fouling leads to a change in pH at the surface of the membrane. Heat dependent materials in spacers can find many applications in MD, such as inducing a change in spacer shape when temperature is lowered leading to membrane cleaning, or removal of scalants from the spacer surface. Indeed, the exploration of smart materials in spacer design provides an exciting avenue for future spacer research.

5. Conclusion

With the end of a period from 2010 to 2021 that saw feed spacers take a more central role in fouling mitigation, to the development and fabrication of numerous novel spacer designs, this review provides a holistic perspective as well as future directions for spacer research. The following are key take-home conclusion points from this review:

- Feed spacers have been gaining prominence since the growth of spiral wound modules (SWMs) as an indispensable component of the SWM. Spacer design should first and foremost address the pressure

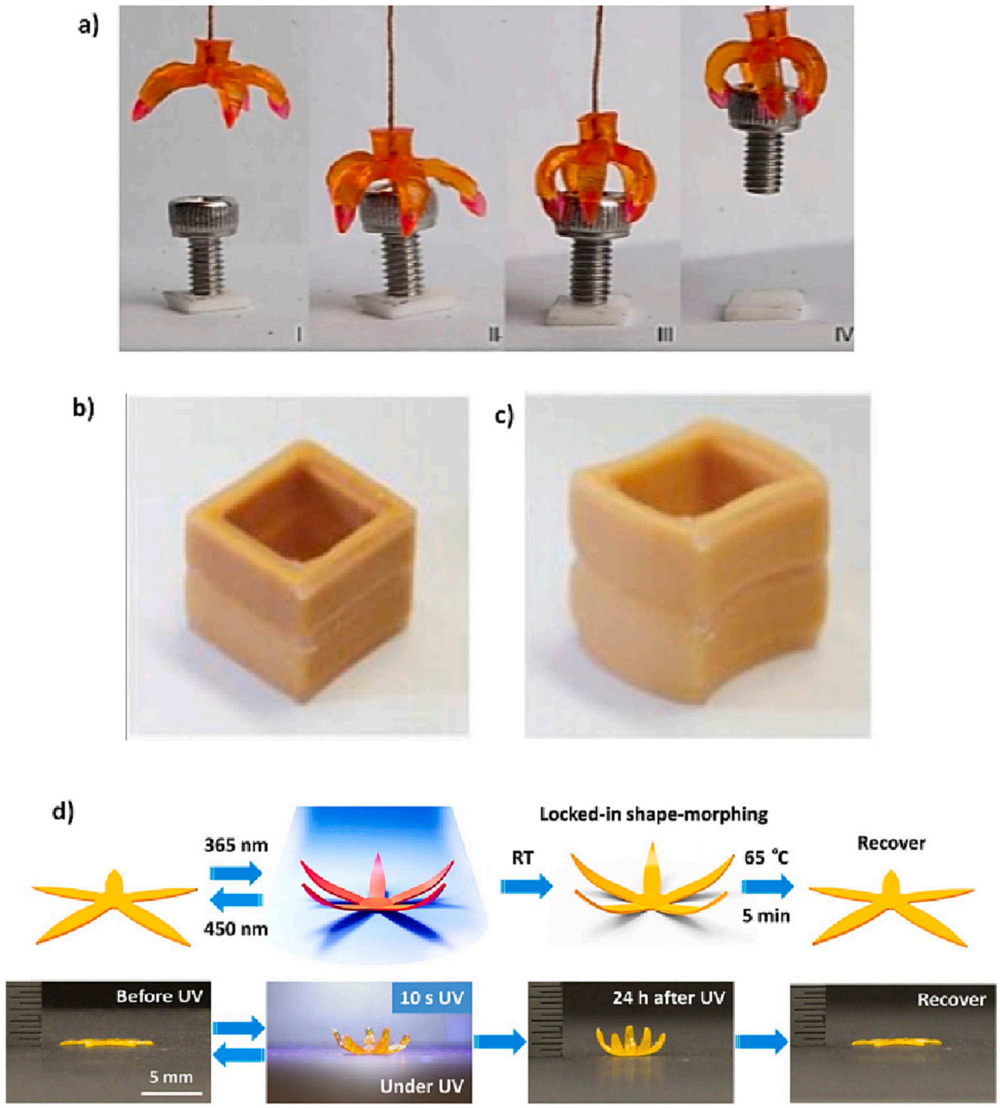

Fig. 15. Examples of 4D printed/smart spacers: **(a)** SMP with thermomechanical properties, that cause the shape to change at varying temperatures [170], **(b,c)** examples of magnetically influenced shape polymers [171], **(d)** elastomers that can switch shape under UV light, and retain the new shape [172].

drop-mass transfer trade-off, by minimizing the energy penalty stemming from generating unsteady flow.

- The role of feed spacers in biofouling development has been well-established. It is generally agreed that spacer filaments and regions with low mixing are points of attachment for foulants, from where they can grow onto the membrane.
- Minimizing the energy demand in pressure-driven membrane processes is desired. The development of new feed spacer designs offers an opportunity to meet this requirement, via FCPD and CP reduction.
- To fully realize the potential of next generation, super-permeable membranes, such as graphene, CNT, and biomimetic aquaporin-based membranes, advanced feed spacers must be developed which can mitigate the severe concentration polarization that comes with super-permeability.
- Given the wide range of applications for which new spacers are developed or modified, a plethora of tools are used to assess spacer performance. These include i) qualitative tools, such as SEM, photography, IR imaging, and particle image velocimetry (PIV), ii) quantitative tools, such as flux drop, rise in TMP, dimensionless numbers analysis, FCPD, and foulant quantifying tools like TOC analysis, and iii) tools that combine both qualitative and quantitative aspects, such as OCT and fluorescence imaging.
- Since 2010 we saw the development of chemical modifications of spacer surfaces, as the role of spacers in biofouling became well established. Coatings that rendered a host of properties to the spacer, from being anti-adhesive to antibacterial, were developed, with varying degrees of success. More recently, we have seen coatings that target organic fouling and scaling as well.
- The development of novel spacer geometries has been of interest for a long time in membrane science, but the past few years saw a huge influx of these designs as 3D printing gained prominence and became widely available. It is important in the coming years that novel spacers are developed with practicality and commercial application in mind. The challenges to the scalability of novel spacer designs include 3D printing technology speed, build size, cost, and material compatibility.
- The development of functionalized feed spacers is forecasted to be a prominent direction of future research. Innovative spacer functionalization can allow it to play a more active role in the membrane process, with functions such as photocatalysis and electrical heating being attempted.
- The utilization of 4D printing and smart materials in spacer design will be an interesting upcoming trend to watch for. With materials that can be modified to be reactive to a host of external stimuli, such

as heat, electricity, pH, etc., we can see a new class of smart spacers that can perform additional roles in the membrane channel.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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