Review

Natural and recycled materials for sustainable membrane modification: Recent trends and prospects

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HIGHLIGHTS

- Membrane modification with natural & recycled materials have been minutely reviewed.
- Harnessing recycled plastic for membrane synthesis has been discussed.
- Natural & recycled materials induce same improvements as common nanomaterials.
- Natural & recycled materials are cost-effective and environmentally friendly.
- Functional groups are important for membrane-fillers interactions.

GRAPHICAL ABSTRACT
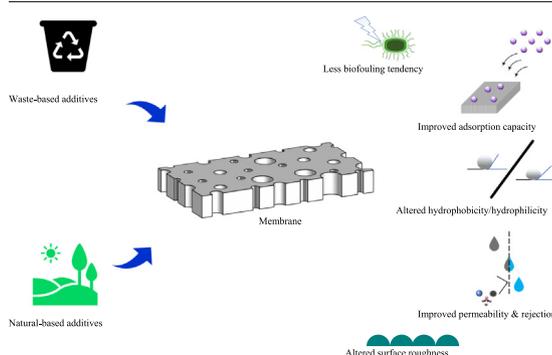
ARTICLE INFO

ABSTRACT

Despite water being critical for human survival, its uneven distribution, and exposure to countless sources of pollution make water shortages increasingly urgent. Membrane technology offers an efficient solution for alleviating the water shortage impact. The selectivity and permeability of membranes can be improved by incorporating additives of different nature and size scales. However, with the vast debate about the environmental and economic feasibility of the common nanoscale materials in water treatment applications, we can infer that there is a long way before the first industrial nanocomposite membrane is commercialized. This stumbling block has motivated the scientific community to search for alternative modification routes and/or materials with sustainable features. Herein, we present a pragmatic review merging the concept of sustainability, nanotechnology, and membrane technology through the application of natural additives (e.g., Clays, Arabic Gum, zeolite, lignin, Aquaporin), recycled additives (e.g., Biochar, fly ash), and recycled waste (e.g., Polyethylene Terephthalate, recycled polystyrene) for polymeric membrane synthesis and modification. Imparted features on polymeric membranes, induced by the presence of sustainable natural and waste-based materials, are scrutinized. In addition, the strategies harnessed to eliminate the hurdles associated with the application of these nano and micro size additives for composite membranes modification are elaborated. The

expanding research efforts devoted recently to membrane sustainability and the prospects for these materials are discussed. The findings of the investigations reported in this work indicate that the application of natural and waste-based additives for composite membrane fabrication/modification is a nascent research area that deserves the attention of both research and industry.

Contents

1. Introduction

Parallel to the steady growth in global clean water demand and the persistent limited availability and stress on these resources, membrane technology is a flourishing research area within the scientific community as a potential tool for water treatment (Wu et al., 2020). Liquid-based membrane processes have established a prominent reputation as powerful technologies for particles and macromolecules separation. Their distinct features compared to conventional technologies are attributed to the potentially low cost, easy processing, and high separation efficiency (Albatrni et al., 2021; Bottero et al., 2006; Zhang et al., 2017a). However, the consistent challenge in maintaining membrane performance for long operational periods has made membrane modification a necessity for prolonged industrial operations under harsh conditions and for treating complex water streams. Based on the separation force, membrane filtration processes are categorised into pressure-driven membranes such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO), osmotic-driven membranes such as forward osmosis (FO) and pressure retarded osmosis (PRO) and thermally-driven membranes such as membrane distillation (MD) (Ladewig and Al-Shaeli, 2017). Each membrane process has a specific molecular weight cut-off and is therefore harnessed for distinct industrial applications. Fig. 1 illustrates the liquid-based membrane process types and their molecular cut-off distribution. The driving forces for each membrane technology category are elucidated along with their capacities for the removal of the common classes of water pollutants. The pure water transfer mechanisms of the different membrane technologies (i.e., exertion of external pressure, salinity gradient, and generation of vapor) are also highlighted. Despite the burgeoning state of membrane technology, the challenges associated with continuous filtration could seriously diminish their full potential. The persistent challenges in these membrane processes include fouling, concentration polarisation, and temperature polarisation (Al-juboori et al., 2021; Li et al., 2019; Wu et al., 2020). Membrane fouling could be defined as the accumulation of undesirable materials on the membrane active layer or in the pores, leading to a permanent or temporary drastic decline in membrane flux, increased resistance to mass transfer, reduced membrane productivity, incurring additional maintenance and operational cost, and shortens membrane lifespan (Al-Shaeli et al., 2021; Ong et al., 2016; Rana and Matsuura, 2010). However, various membrane modifications have been suggested as an effective way to cope with these limitations.

This research field of membrane modification has progressed drastically in recent years. Membrane modification is defined as revising the surface characteristics of the membrane surface to impart the attractive properties of a material at its nanoscale into the membrane structure while maintaining the intrinsic bulk properties of the membranes (Kochkodan and Hilal, 2015; Ladewig and Al-Shaeli, 2017). This can be achieved via versatile surface (e.g., surface functionalization and/or post polymerization), and bulk modification routes. It is generally believed that enhancing the membrane's hydrophilic nature could influence the interactions between foulants and membrane and thereby, improve the entire performance of the target membrane. A wide spectrum of chemical and physical routes have been devoted to this goal (Al-Shaeli et al., 2021; Al-Shaeli et al., 2017). Within the past two decades and with the emergence of nanotechnology approaches, the application of nanoscale materials for membrane modifications became a hot research area. A very large number of inorganic, organic, and organic-inorganic frameworks have been harnessed to produce so-called "nanocomposite membranes" (Siddique et al., 2021). Nanocomposite membranes not only merge the characteristics of nanomaterials and organic polymer but could also manifest novel desired features, such as hydrophilicity, porosity, improve membrane performance, and improved mechanical strength (Al-Shaeli et al., 2021; Zeng et al., 2022). However, there is a very active discussion questioning the economic feasibility and environmental fate after the service life of commonly applied additives (DeFriend et al., 2003; Yoshino et al., 2005). Hence, there is a constant effort to employ alternative materials with sustainable features for membrane modification.

Naturally occurring and waste-based materials, such as clays, Arabic Gum, aquaporin, lignin, biochar and fly ash, have extensively been
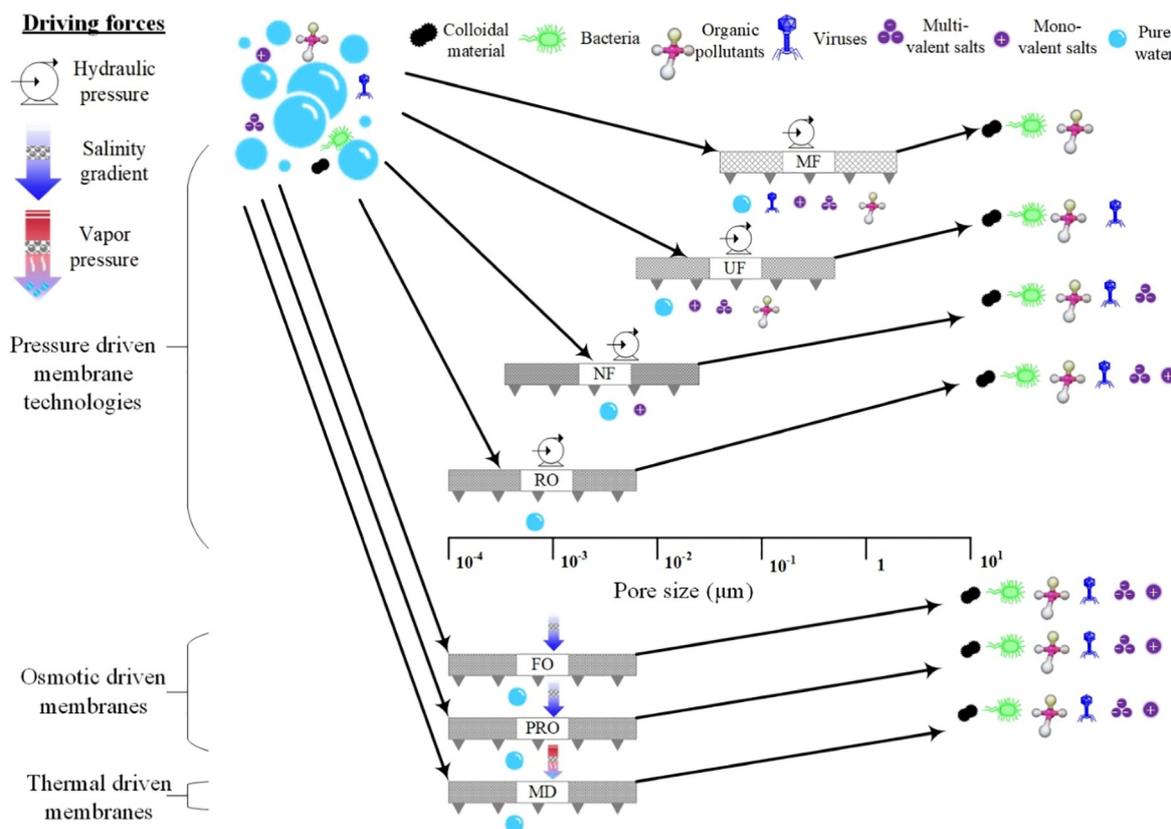
Fig. 1. Membrane filtration processes categorisation based on their driven force and their molecular cut-off.

employed as adsorbents for water treatment applications (Maged et al., 2020). These special considerations, regarding the flourishing of these materials, were attributed to their hydrophilic nature, abundance, cost-effectiveness, and environmental sustainability (Bahi et al., 2017; Rosales et al., 2017; Yang et al., 2019). In particularly, they require only mild chemical processing compared to other common additives (Farahani and Vatanpour, 2019). Also, utilizing recycled materials could be beneficial from a waste management perspective. As part of this recent research trend aiming to fulfill sustainable development and circular economy goals, harnessing these additives for membrane modification applications could be a step forward considering the "Green nanocomposite membrane" approach. Natural and waste-based additives have repeatedly been reported to bestow promising enhancements onto polymeric membranes characteristics. Their integration within the membrane's structure could induce desired changes to the physicochemical properties and antifouling features (Buruga et al., 2019). This will in turn reflect on the final membrane permeation/retention characteristics. However, as with many other additives, these adsorbents generally show inevitable weak interactions with polymeric chains and may necessitate certain processing to improve that compatibility (He et al., 2016; Zaidi et al., 2019).

Herein, this review will shed the light on versatile sustainable materials, including natural, recycled, and recycled waste additives, utilized for membrane modification purposes. Also, a detailed discussion pertaining to the limitations of these materials and possible modification methods are presented. In particular, the literature illustrated the potential of plastic waste as a polymeric materials source. Ultimately, prospects and challenges have been reported. Concentricity was made about four criteria throughout the whole manuscript, namely membrane performance, economic feasibility, environmental and health concerns, to probe and validate the potential of these additives against common ones. Future perspectives and authors' insights are given to cope with current limitations. There are few review papers that focused only on natural additives such as those reported on the application of montmorillonite (Dlamini et al., 2019) and Aquaporin (Tang

et al., 2015). However, to the authors' knowledge, there is no review covering the application of both natural and waste-based additives in the preceding literature yet. The following sections enlist and describe the versatile natural and recycled waste additives reported to date. Besides, the observed structural changes of the membranes and their performance enhancements induced by these additives are critically discussed.

2. Sustainable natural additives for membrane modification

Natural additives are defined as materials that could be obtained from natural sources. These additives have been utilized directly without further treatment or with a mild structural modification. Many natural materials suitable for membrane modification are abundantly available. These materials demonstrated their promising potential in a wide spectrum of applications for water and wastewater treatment such as adsorption, coagulation, and sedimentation. This distinguished potential to remove contaminants has attracted the scientific community to harness them for the fabrication/modification of liquid-based membranes. Natural materials were harnessed to minify the nanocomposite membrane fabrication cost while imparting the desired characteristics. For instance, naturally-occurring Kaolin clays were employed for ceramic microfiltration applications (Rekik et al., 2016), apatite was used for microfiltration and ultrafiltration membranes applications (Masmoudi et al., 2007), and fly ash was employed for preparing low-cost MF membranes (Singh and Bulasara, 2015). These natural additives have been utilized as raw materials without further modification while sometimes modification is required to bestow greater compatibility and homogeneous structure, as will be illustrated in the subsequent sections.

2.1. Clays

Clays are natural hydrophilic materials with a primary component of tetrahedral $[SiO_4]^{4-}$ and octahedral $[AlO_3(OH)_3]^{6-}$ sheets

(Mukhopadhyay et al., 2019). With crystalline structures, clays exhibit a particle size of less than 2 μm (Uddin, 2017). These natural materials have been intensively utilized for aqueous solution treatment processes such as adsorbents, membrane materials, and additives for mixed matrix membrane fabrication. Success in these applications has been attributed to their high aspect ratio, chemical and mechanical stability, micro porosity, high adsorption capacity, layered structure, simplicity to prepare and scale-up, and low cost (Asadi and Zerafat, 2016; Mukhopadhyay et al., 2017; Sarkar et al., 2013; Sarkar et al., 2012). Other forms of clay such as kaolinites, iolites, sepiolite, montmorillonite, and attapulgite, have been used as additives in the casting solution to enhance the performance of membranes (Buruga et al., 2018). Montmorillonites (Na-MMT) and Cloisite were largely utilized as nanofillers in polymeric membranes and have revealed an outstanding enhancement in the thermal and mechanical characteristics of the produced composites along with better performance (Guo et al., 2013; Lakshmi et al., 2008; Shokri et al., 2021; Simona et al., 2017). However, the application of natural clay minerals is restricted due to their limited potential in eliminating micro contaminants from water owing to their pore size and regeneration requirements after exhaustion (Bhattacharyya and Gupta, 2008; de Paiva et al., 2008; Sarkar et al., 2019; Unuabonah and Taubert, 2014; Unuabonah et al., 2018). Additionally, clays particles manifest a lower affinity toward organics as the hydration of the clay surface reduces the chances of organic materials penetration into clays' layers (Borisover et al., 2008). Therefore, researchers have developed clay-polymer composite membranes to overcome these drawbacks and enhance the characteristics of polymeric membranes (Rodrigues et al., 2019).

Commonly, there are three methods for clay-polymer composite membrane fabrication, including; solution blending, in-situ polymerization, and melt blending (Dlamini et al., 2019). The solution blending method entails the preparation of a homogenous mixture of clay-polymer-solvent and casting in a non-solvent environment. The polymer harnessed should be water-soluble (Billingham et al., 1997). In-situ polymerization involves the initial functionalisation of monomers with clay minerals before polymerization. Homogenous dispersion of the clay within the polymeric matrix is crucial to obtaining a unique nanocomposite structure (Dlamini et al., 2019). The melt blending technique includes the physical mixing of polymers and clay minerals, followed by compounding and then annealing processes via mixers/extruders. Although this method is regarded as simpler and more environmentally friendly, it is generally used for hydrophobic thermoplastic polymers which are not suitable for water treatment applications (Dlamini et al., 2019). Clays can interact with polymers in three different forms: microscale clay, nanoclays, and organoclays. Microscale clay is usually used for synthesis and modification of inorganic membranes. Nanoscale clay or nanoclays are nanoparticles of layered mineral silicates with a diameter of 50–200 nm and nanometric thickness of 1 nm (Ghormade et al., 2011; Wilson, 2003). They have a high surface area, well-dispersed, have higher thermal stability and chemical resistance, and good reactivity compared to micro clays (Ghormade et al., 2011; Leszczyńska et al., 2007; Wilson, 2003; Yu et al., 2004). They are usually blended with polymers to form nanocomposite membranes which consequently improves the mechanical and thermal properties, improve performance, and anti-fouling properties of the membranes (Maiti et al., 2008). However, a low content of nanoclays (less than 10 wt%) is required to assure the desired effect. Organoclays are clays that are modified with different modifying agents such as surfactants, coupling agents, organic materials, polymer and ionic liquids to improve mechanical properties, the adsorption capacity of clays and reduce the immiscibility between hydrophilic clay particles and hydrophobic polymeric membranes (Chansuvarn, 2017; Liu et al., 2011). Organoclays can be used to remove heavy metals ions from aqueous solutions (Jang et al., 2020).

Clay-based nanocomposite membranes have received considerable attention due to their multifunctionality. Merging the clays with polymeric membranes matrix is a recent hot research area to endow an ecofriendly membrane with self-cleaning features and enhanced performance. A wide spectrum of applications was reported in the literature, such as pervaporation (Choudhari and Kariduraganavar, 2009), greywater treatment in moving bed reactors (Rakovitsky et al., 2016), fuel cells (Plackett et al., 2011), separation of sodium dodecyl sulfate (Workneh and Shukla, 2008) and membrane distillation (Prince et al., 2012). Clays could be harnessed for both surface and bulk modification. Literature has repeatedly reported that the addition of clay increases membrane porosity and hydrophilicity (Wang et al., 1998). Several studies consistently reported that the amalgamation of clay with polymeric membranes enhanced their structural stability, however, this depends on the cohesion between clay and polymer. When the polymer chains interact with the clay functional groups such as -OH groups in the silicate layer, this results in a mechanically robust structure (Dlamini et al., 2019). Clay minerals have been also reported to enhance the crystallization of polymers (Sharma et al., 2011). In general, it should be noted though that the mechanical structure improvement does not hold true for a high concentration of additives. The higher clay concentration reduces the distances amongst them and consequently promotes the agglomeration induced by van der Waals forces. This effectively makes them a weakness point in the composite structure (Dlamini et al., 2019). Meanwhile, raising the percentage of the added clay to the composite could also increase the viscosity of the casting solution which has been linked to the reduction of water diffusion (Dlamini et al., 2019). Therefore, there are important key aspects that need to be considered when adding clay particles to membranes. These include selecting an appropriate loading percentage that strikes a good balance between improving membrane characteristics and maintaining its durability while ensuring a strong physical interaction between clay particles and polymer. This may require chemical modification of either component, adding supporting agents, and selecting a suitable casting method. Table 1 showcases an overview of recent attempts of using natural clays for nanocomposite membrane applications.

Modification of clays is extensively practised by many researchers as it preserves the original properties of clays and introduces new properties such as hydrophobicity, hydrophilicity, anti-static properties, and biocompatibility (Buruga et al., 2019). This offers an effective pathway to optimise different parameters (e.g., swelling properties, adsorption properties, and rheological/colloidal characteristics of the clays) for proposed uses. Clays can be modified with different materials, such as cationic surfactants, gemini surfactants, and manganese oxide to improve the adsorption capacity and the mechanical strength of clay (Chansuvarn, 2017; Liu et al., 2011). Different methods have been used for this purpose including reaggregation, calcination and delamination, adsorption, dihydroxylation, inorganic, and organic anions integration into clays' structure. Furthermore, inorganic cations and cationic complexes, ion exchange with organic cations reaction with acids, pillaring with poly-cations, grafting of organic compounds, and inter and intra-particle polymerization have been also reported for clay modifications (Bergaya and Lagaly, 2001). Apart from that, physical techniques were also used for clay modifications such as ultrasound and plasma (Kalıpcı, 2019; Şahin et al., 2015).

An example of the common modifications practiced in synthesising clay-polymer composite membrane is the replacement of metal cations between the clay interlayers with organic cations to enhance the interaction between clays and polymer (Dlamini et al., 2019). The modified clay in this form is commonly termed organo-clay or nano-clay. This alteration in clay structure is normally applied to reduce the immiscibility between hydrophilic clay particles and hydrophobic polymeric membranes. The replacement of exchangeable ions in the clay layers may increase the interlayer spacing and this could improve water flux, but at the expense of the selectivity of the membrane (Dlamini et al., 2019). Prince et al. (2012) prepared a highly hydrophobic PVDF–clay nanofiber membrane for desalination application using direct contact membrane distillation. With clay percentages of 4% and 8% (wt%), the salt rejection was maintained higher than 99% for the 8 h operation, whereas the rejection of the neat membrane recorded was ≈ 97% for the same operation time. For forward osmosis, membrane application, polyvinyl alcohol and functionalised montmorillonite clay were prepared and employed as membrane substrate for constructing a separating layer of polyelectrolyte via the layer-by-layer method (Pardeshi and Mungray, 2014). Compared to the

Table 1
Recent studies of clay-based modified membranes used for water treatment applications.

Additives	Membrane type and materials	Feed	Altered membrane feature	flux	rejection	Ref
China clay	MF (aluminium fluoride trihydrate and aluminium oxide	Synthetic Oil/water emulsion	Increased flexural strength, high pure water permeance, and chemical stability	Flux achieved with this membrane was 200 LMH	96% with oil concentration 200–1000 mg/L	(Rashad et al., 2021)
Clay nanoparticles (CN)	UF (PSF)	10 mg/L sodium alginate solution	Improved thermal and mechanical stability, increased porosity, hydrophilicity, and surface charge.	Increased relative permeability from 68.6 LMH/bar for PSF to 73.4 LMH/bar after the addition of 1.5 wt% of CN	Increased rejection by 0.5%	(Rodrigues et al., 2019)
Montmorillonite (MMT) Montmorillonite (MMT)	Polysulfone nanocomposite (PSF)	–	Good dispersion of MMT was achieved. Noticeable improvements in the hydrophilicity, morphology, and mechano-thermal properties of the modified PSF membranes	No evaluations regarding water treatment performance were conducted	–	(Anadão et al., 2010)
Single platelet MMT with and without sodium hexametaphosphate single platelet montmorillonite	PES UF membranes	Ultra-pure water, sodium alginate solution, and natural surface water	Higher membrane performance was achieved. The modified membranes with clays had a higher susceptibility to fouling in comparison with the control membranes.	The achieved permeance was 312 LMH/bar and 389 LMH/bar when the addition of clay was 1% and 2% as compared with pristine PES (176 LMH/bar)	Alginate rejection was 97.6 ± 0.3, 96.9 ± 0.3, when the concentration of clay was 1% and 2% while pristine PES achieved 97.7 ± 0.3	(Mierzwa et al., 2013)
Halloysite clay nanotubes (HNT) modified with platinum or palladium (pt, pd) NPs	Cation exchange membrane (tetrafluoroethylene and per fluorinated sulfocontainning monomers)	NaCl solution	Increased hydrophilicity.	Improved diffusion permeability by about double.	–	(Petrova et al., 2019)
N-Halamine grafted halloysite nanotubes (N-halamine@HNTs)	PES membranes	Synthetic water consists of water and PEG 20KDa	Increased storage modulus and pores size. Reduced hydrophobicity and surface roughness. Improved anti-bacterial activity against *E.coli*.	Increased pure water flux by more than 5 folds at a loading of 1 wt% N-halamine@HNTs. However, increasing the loading to 3% only increased the flux by more than 3 folds. Increased	90% rejection of PEG (2000KDa) when the concentration of 1 wt% of N-halamine@HNTs reduced PEG rejection from ca. 95% to ca. 75%. Further increase of loading to 3% rose the rejection to approximately 90%.	(Duan et al., 2015)
Cloisite 30B modified with organosilyl groups (vinyl (Cloisite30B-Vy), octyl (Cloisite30B-C8), and octadecyl (Cloisite30B-C18))	PES electrodialysis cell membrane	synthetic zinc solution (10 g/L)	Increased thermal stability, hydrophobicity, and polarity.	All composites increased the flux. Cloisite30B-C8 resulted in the highest increase of approximately 16%.	All composites increased the flux. Cloisite30B-C8 resulted in the highest increase of approximately 16%.	(Simona et al., 2017)
Mt modified with poly (4-styrene sulfonic acid-*co*-maleic acid) (PSSMA)	PVA on a porous PSf support material	Salt solution (Na_2SO_4) and pharmaceutical synthetic solutions of Cephalexin Amoxicillin And Ibuprofen	Higher hydrophilicity and roughness properties.	Increased pure water flux by about 17.5% for Mt-m loading of 0.4 wt%	Na_2SO_4 Rejection only decreased from 95.75% to 94.75% for Mt-m loading of 0.4 wt%. Cephalexin and Amoxicillin rejection did not change with Mt-m incorporation, Ibuprofen rejection decreased.	(Medhat Bojnourd and Pakizeh, 2018)
Cloisite (CS)-15A modified with dimethyl dihydrogenated tallow quaternary ammonium salt	Polyamide (PA) RO membranes	Saline water (DI + NaCl)	Slightly increased hydrophobicity and surface roughness.	CS-15A loading percentage of 0.02 wt% slightly increased flux, but lower percentages decreased flux slightly.	CS-15A loading percentage of 0.02 wt% and above slightly increased rejection.	(Zaidi et al., 2019)
Montmorillonite MMT-Fe_3O_4, MMT-HBE, and MMT-acid Activated (AA)	PES mixed matrix nanofiltration membranes	Salty solutions (NaCl, Na_2SO_4 $MgSO_4$ and dyes (methyl orange MO;	All additives increased roughness, hydrophilicity, porosity, and mechanical strength (for low filling load).	Pure water flux increased by around 250% for Mt-AA, 400% for Mt-HBE, and 450% for Mt-Fe_3O_4	All modification approaches with their different filling load improved the rejection of dyes, heavy metals	(Gozali Balkanloo et al., 2020)

Table 1 (*continued*)

Additives	Membrane type and materials	Feed	Altered membrane feature	flux	rejection	Ref
		methylene Blue MB) and BSA	Mt-Fe$_3$O$_4$ reduced negative surface charge, but the other modifiers increased negative surface charge. All modification approaches led to improve fouling resistance of the membrane.		(Zn, Ni, Cu, and Cd), and various salts (NaCl, Na$_2$SO$_4$, and MgSO$_4$)	
Montmorillonite modified with folic acid (Mt-FA)	PVC UF membranes	Humic acid solution	Higher membrane hydrophilicity and surface roughness were achieved. Better mechanical strength and higher porosity were obtained at a loading percentage up to 1.5 wt %of modified Mt-FA. All loading percentages decreased the irreversible fouling ratio, but the reversible fouling ratio increased for all loading percentages except for Mt-FA 2 wt%.	Flux increased by a maximum of about 65% with Mt-FA loading of 1.5 wt%	Rejection in general increased with all the filler loading. The highest rejection increase of more than 400% was achieved with Mt-FA loading of 1 wt%. The membrane also showed good rejection for humic acid and Pb.	(Shokri et al., 2021)
Mg-Fe layered double hydroxide modified montmorillonite (LDH-Mt)	Polysulfone/Polyvinylpyrrolidone (PSF-PVP) UF	Oily water mixture (transformer oil and motor oil)	Higher hydrophilicity, thermal stability, and membrane roughness.	Pure water flux increased by roughly 200%. Oily water flux increased by about 75%.	BSA rejection increased by about 50%. Sodium Lauryl Sulfate rejection for transformer and motor oils increased by 21% and 9%, respectively. Acetyl trimethyl ammonium bromide rejection for the respective two oils was 11% and 7%.	(Makwana et al., 2020)
Mt nano clays	electrospun nanofibrous membranes (PVA/chitosan ENM)	Basic blue 41 (BB41) dye solution	Increased mechanical strength, pore size and porosity.	Pure water and colored wastewater flux increased with increasing the filler loading. Maximum flux increases for both pure and pure and colored water reached 60% with filler loading of 3 wt%.	Dye removal decreased by about 28% with filler loading increase from 0 to 3 mass %.	(Hosseini et al., 2019)

commercially available HTI membrane which has a flux of 13 LMH, a higher water flux was achieved (37.65 LMH and 25.5 LMH) for AL-DS and FO, respectively. Besides that, an impressive reverse salt diffusion of 0.2813 gMH was reported in the active layer facing draw solution (AL-DS) and 0.0510 gMH for FO. As claimed, this noticeable improvement was ascribed to the enhanced porosity and thermal stability of the composite membrane upon the addition of clay. Likewise, Saffaj et al. (2006) prepared UF membranes using a clay support layer, a zirconia mid-layer, and a TiO$_2$-ZnAl$_2$O$_4$ top layer with a 5 nm pore diameter. The modified membrane was evaluated for filtering a mixture of NaCl and CaCl$_2$ salts and methylene blue and orange acid dyes. With these low-cost local natural materials, Saffaj and co-workers achieved a high rejection for heavy metals (>87%) and dyes such as orange acid (≈ 90%) (Saffaj et al., 2006). Modified montmorillonite via lysine amino acid was harnessed for MMMs fabrication by Shokari and Yegani (Shokri and Yegani, 2017). The batch adsorption test of the modified membranes as a function of solution pH and arsenic initial concentration was conducted to assess the performance of the modified membranes. The authors reported that the arsenic adsorption was most favourable at neutral pH while the reusability of the nanocomposite was good even after five cycles of adsorption-desorption experiments in dead-end filtration with surface water (Shokri and Yegani, 2017). The authors applied a simple alkaline wash (i.e. water at pH = 9)

and was sufficient to effectively restore membrane adsorptivity. This may indicate the capacity of additives to induce anti-fouling properties in the prepared MMMs especially toward inorganic foulants.

The two common clays, Bentonite and Kaolin will be discussed in separate sections due to their extensive applications in membrane manufacturing and modification as compared to other types of clays. Zeolite will also be addressed in a separate section due to its distinctive differences from clays. Clay minerals are two-dimensional layer structures with exchangeable cations and a variable amount of water in the interlayer region. In clay minerals, for instance, smectite, the attractive forces between the individual layers are almost balanced by expansive forces due to the hydration of the internal cation. Water enters the interlayer region to interact with the interlayer surface and then hydrates the interlayer cations (Bish, 2006). Zeolite, in contrast, has a three-dimensional layer structure connecting aluminosilicate framework structure and their frameworks. These frameworks are typically organised in three-dimension form and are formed in large structural cavities in which water exchangeable cations occur (Campbell and Cheetham, 2002). Structural studies have revealed that water interacts with both the exchangeable cations and the framework, and water usually enters the extra framework sites progressively as the partial pressure of H$_2$O is enlarged (increase in relative humidity) (Bish, 2006).

2.2. Bentonite

Bentonite (Bent) is a naturally abundant clay that belongs to the smectite group. This group has an arrangement of aluminium-phyllosilicate ($Al_2O_3.4SiO_2. H_2O$) with an octahedral alumina sheet and two layers of tetrahedral silica sheets as shown in Fig. 2. As illustrated, a single structural unit of bentonite consisting of an aluminium octahedral sheet is layered between two tetrahedral silica sheets (Gao et al., 2016; Hebbar et al., 2016). Bentonite is an impure ore of clay, which constitutes mainly montmorillonite and is obtained from volcanic ashes (Chen et al., 2014; Dutta et al., 2021). It has a permanent negative charge on the structure lattice emanating from the substitution of the silicon ions with ferric or aluminium cations in the tetrahedral sheets, and the exchange of aluminium ions with ferrous or magnesium cations in the octahedral sheets (Hu et al., 2006; Kapoor and Viraraghavan, 1998). It is noteworthy that bentonite has a pH-dependent negative or positive charge at the edges. Bentonite sustains charge neutrality with the cations present in the interlayer between the crystals such as Ca^{+2} and Na^+. In aqueous environment, these cations can exchange with other cations surrounding them (Worral, 1968).

Bentonite has extensively been harnessed in a wide spectrum of applications including cosmetics, catalyst, polymer filler, bacterial remediation (Nidhi Maalige et al., 2019; Santos et al., 2011; Vernhet and Bellon-Fontaine, 1995), treatment of oily wastewater (Alther, 1995; Gitipour et al., 1997; Moazed and Viraraghavan, 2005; Okiel et al., 2011), removal of dyes from wastewater (Ramakrishna and Viraraghavan, 1997; Tahir and Rauf, 2006; Toor and Jin, 2012) and pharmaceuticals due to its physicochemical properties such as small particle size, high surface area, high cation exchange capacity, high porosity, and swelling property (Doulia et al., 2009; Haider et al., 2017). It has been used in the ion exchange process and as potential adsorbent material to adsorb heavy metal ions (e.g. Cu^{+2}, Cd^{+2}, Ag^{+2}, Zn^{+2}, Pb^{+2}, Cs^{+2}, and Cr^{+2}) from aqueous solutions (Ayari et al., 2005; Katsou et al., 2011; Pandey, 2017; Prabhu and Prabhu, 2018; Schütz et al., 2016; Taha et al., 2016; Triantafyllou et al., 1999; Yadav et al., 2019). Although bentonite can adsorb metal ions, its uses as an adsorbent in filtration mode are restricted in practice due to two reasons, (1) bentonite's propensity to swell upon exposure to water forming suspension that is hard to separate, and (2) the compactness of bentonite particles in column configuration leads to low water permeability (Abdukhalikov, 2017; Kapoor and Viraraghavan, 1998). The adsorptivity of bentonite is attributed to its capacity to exchange the cations in the interlayer of the crystals with those in the surrounding environment (Dutta et al., 2021). Many research articles demonstrated that modifying bentonite into organo-bentonite improves its capacity to remove organic pollutants. (Bae et al., 2000; Smith and Galan, 1995; Zhu et al., 1997). This modification is conducted by replacing the interlayer cations with quaternary amine cations.

Bentonite has been harnessed as nanofillers in polymeric membranes for water treatment applications due to its attractive features, including low cost, availability, high hydrophilicity, high surface area, and aspects ratio, net negative charge, good compatibility with polymer material, facile chemical and modification ability (Hebbar et al., 2018). Bentonite is capable of enhancing membranes' mechanical strength even at very low loading percentages (Singhal and Datta, 2007), meanwhile, it could bestow desirable traits on
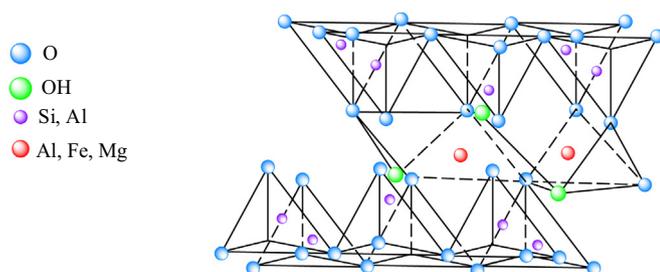

Fig. 2. Bentonite structure reproduced from (Bananezhad et al., 2019). Used with copyright permission from Elsevier.

membranes' surfaces, including higher hydrophilicity, greater rejection, higher permeation flux, and improve the anti-fouling features (Dechnik et al., 2017; Kadhom and Deng, 2019; Kumar et al., 2015; Mondal et al., 2017; Noble, 2011). The presence of negative bentonite particles in the composite membrane can endow promising properties for wastewater treatment applications due to the synergistic effects of electrostatic interactions and cation exchange capacity (Abollino et al., 2003; Kraepiel et al., 1999; Mercier and Detellier, 1995). However, alongside these advantages, the introduction of bentonite and other clay materials as inorganic filler for membrane process applications is limited to relatively few examples in the literature (Ahmad et al., 2018a; Ahmad et al., 2018b; Ghaemi et al., 2011; Gozali Balkanloo et al., 2020; Hebbar et al., 2014; Kumar et al., 2015). This is due to its restricted adsorption capacity which limits its practical advancement (Khenifi et al., 2007). Therefore, it is critical to modify Bentonite with some functional groups to enhance the adsorption capacity (Özcan et al., 2007).

Panpanit and Visvanathan (2001) explored the feasibly of a membrane modified with natural bentonite clay for oily water separation. The study concluded that natural bentonite clay incorporation into UF membrane structure could diminish the adsorption layer on the membrane surface and internal pores. In another work, bentonite incorporation into a membrane matrix was capable of enhancing the removal efficiency of hazardous heavy metals from water due to their highly porous surface (Kapoor and Viraraghavan, 1998). Guo et al. (2019) employed modified bentonite with polyene propylamine to remove anionic dyes, Amido black, from wastewater. The obtained results demonstrated that the maximum adsorption capacity of dye by Bent-PAA was 144.08 mg/g at pH = 2 and contact time = 120 min, compared to nearly zero at different pH ranges for the unmodified Bent. In another work, Hebbar et al. (2018) fabricated a low-cost nanocomposite membrane by grafting poly (4-styrene sulfonate) brushes on bentonite using the distillation-precipitation polymerization method. The modified bentonite was then added to a mixture of PEI and PVP through the immersion precipitation method. It was reported that the resultant membranes displayed a significant enhancement in surface hydrophilicity, porosity, and water uptake capacity at 3 wt% modified bentonite compared to unmodified membranes. Besides, the water flux was 211 LMH compared to the unmodified membranes (121 LMH) along with greater antifouling characteristics where 75.6% of fouling was reversible. In another study, Nidhi Maalige et al. (2019) prepared low-pressure nanofiltration membranes utilizing unmodified and modified bentonite (sulfonated Bent S-BEN). Bentonite particles were impregnated within the polyamide layer (PA) via the interfacial polymerization method. The modified/S-BEN-PA membranes showcased 2–3 times higher water flux and higher rejections against $MgSO_4$ salt (>20%), malachite green oxalate dye (>98%), and humic acid compared to nascent PA and BEN/PA membranes. EDTA functionalised bentonite clay particles were harnessed by Dutta et al. (2021) to prepare PSF-MMMs. The functionalised bent turned the MMMs surface charge highly negative which facilitated the adsorption of heavy metals as confirmed by zeta potential measurement. Results demonstrated a greater self-cleaning feature for the modified membranes due to the enhanced hydrophilicity whereas it showed the highest permeability of 7.4×10^{-11} LMH/bar. Removal of metal ions was in the range of 92–98% for Ni^{+2} Cu^{+2}, Zn^{+2}, and Pb^{+2}. In other research, a TFN FO membrane was prepared by impregnating bentonite nano clay into PVDF electro-spun nanofiber supported polar crystalline phase to improve the performance of the membrane (Shah et al., 2020). The modified FO membranes disclosed a significant improvement in their performance. A greater water flux (40.64 LMH at 1 M NaCl draw solution) was achieved for TFN membranes when the highest loading content of bentonite nano clay was used with a barely tangible increase in reverse solute flux (RSF). Table 2 summarises the outcomes of recent studies concerning the use of bentonite as fillers in membrane technology.

2.3. Kaolin

Kaolin is another naturally occurring clay type. It is mainly composed of kaolinite and small amounts of quartz and mica (Zhu et al., 2010). The

Table 2
An overview of up-to-date studies on bentonite incorporation into membrane technology.

Membrane type and materials	Feed type	Altered membrane features	Flux	Rejection	Ref
PES and CA membranes	Car wash effluent	Better membrane performance for CA/SPEEK/Bent reflected in higher flux and flux recovery ratio compared to commercial PES membranes	102 LMH for CA membranes and 95 LMH for modified PES membranes	60% COD removal for CA membranes and 47% COD removal for PES membranes	(Kiran et al., 2015)
Different types of clay (bentonite, sepiolite, and zeolite), MF polysulfone (PSf)	Cu (II) solution with a concentration of 5 mg/L	Increased hydrophilicity, porosity, and negative surface charge.	Increased water flux from almost none for PSF to a maximum of 140 LMH for sepiolite/PSf	Increased Cu (II) removal from $\approx$ 0 for PSF to $\approx$ 95% for PSF/zeolite	(Abd Hamid et al., 2020)
PVDF, PEI, PES, PAI, PPSU, and CA	dairy wastewater	Water permeation flux was higher for membrane PES/SPEEK/Bent as compared with other polymeric membranes. A lower contact angle (54.05°) was achieved for CA/SPEEK/bentonite Membranes.	Water permeability with this composite 66.66×10^{-9} LMH/bar	–	(Pagidi et al., 2015)
UF (PSF)	Synthetic oil-water emulsion	Increased roughness by about 5 times, decreased contact angle by $\approx$ 40% when loading of 10 wt% was used, increased porosity by a maximum of $\approx$ 3 times, and pure water permeability by $\approx$ 5 times with loading of 8 wt%.	Increased water flux by a maximum of about 6 times with loading of 8 wt%.	A slight decrease in oil rejection by about 3% loading of 8 wt%.	(Kumar et al., 2015)
Thin-film nanocomposite membranes (TFN)	Saline/brackish water (2000 mg/L) of NaCl solution	Higher membrane hydrophilicity was achieved when small amounts of Bent NPs were added, and higher membrane performance was obtained for all the TFN membranes.	45.6 for TFC to 58.8 LMH For TFN membranes after added 0.15 wt% of Bent NPs	96.25% for TFC to 97.3% to TFN after added 0.15 wt% of Bent NPs	(Kadhom and Deng, 2019)

theoretical composition of the kaolinite is $(Al_2.Si_2O_5.(OH)_4)$ with 39.50% Al_2O_3, 46.54% SiO_2, and 13.96% H_2O (Bhattacharyya and Gupta, 2008). Kaolinite has a unique structure that consists of two layers. The interspace layer of the structure is covered with octahedral sheets (hydroxyl groups of Al_2O_6) while the other side of the structure is covered with oxygen molecules of SiO_4 tetrahedra, as shown in Fig. 3. These layers are bonded together by hydrogen atoms and van der Waals forces to obtain a distinct space between the layers which is different from other types of clays (e.g. MMT and bentonite) (Babu Valapa et al., 2017). Kaolinite was chosen as a modification material because of its outstanding properties including high-temperature stability, high chemical resistance, easily moulded, high crystallinity, fine texture, and fine grain size of 0.2–1 μm. It is a soft material and has a low viscosity at high solids contents in many systems, is easily dispersed and wetted in water and some organic solvents, and can be formed with a controlled particle size distribution (Bellotto et al., 1995; Breck, 1984; Klein, 1985). Since it is a cheaply available material, it has been used in membrane technology as a filtration layer or as a support layer due to its mechanical properties, good adsorption properties, and pore structures that can be achieved through thermal processing (Vimonses et al., 2009). It has been widely used in applications of inorganic membrane (ceramic membranes) synthesis as support layers (Boudaira et al., 2016;

Fig. 3. Kaolinite structure reproduced from (Babu Valapa et al., 2017). Used with copyright permission from Elsevier.

Harabi et al., 2014; Zhou et al., 2010) and active layers for microfiltration (Hedfi et al., 2014; Jana et al., 2010; Mohtor et al., 2017) and ultrafiltration (Ben Ali et al., 2018).

Kaolin is considered as a more attractive additive for synthesising low-cost modified membranes compared to the common nanomaterials such as TiO_2, Al_2O_3, and ZnO (Marino et al., 2017). Particularly, kaolin was the second most used alternative material for synthesising inorganic membranes (Abdullayev et al., 2019). From scrutinising the literature, one can notice that there is a lack of studies impregnating natural kaolin within polymeric membranes while it has commonly been studied as an additive or main structural material for constructing inorganic membranes (Belgada et al., 2021). It is unsuitable as a standalone material for ceramic porous membranes due to its poor sinterability, resulting in a weak mechanical structure of the sintered membrane matrix(Chen et al., 2017). Recently, Belgada et al. (2021) have employed kaolinite as an additive for manufacturing phosphate-based microfiltration membranes. The study found that membranes produced at optimum conditions of 15% kaolinite with a sintering temperature of 1000 °C for 2 h exhibited higher performance in treating textile wastewater with turbidity, TOC, COD, and BOD removal rates of 99%, 69.4%, 74%, and 77.1%, respectively. Similarly, unmodified kaolin application for polymeric membrane modification poses some limitations due to their weak interactions with the polymeric chains. Therefore, kaolin modification becomes a necessity to enhance the interactions with the polymers and strengthen the mechanical properties. Liu et al. (2019) fabricated a low-cost 2D membrane by incorporating 2D kaolin nanosheets prepared by physical and chemical exfoliation and laid onto a support layer made of CA (see Fig. 4). The CA/kaolin membrane exhibited an ultra-high permeability (~4000 LMH/bar) which was about 8 times higher compared to a commercial PES membrane (500 LMH/bar). This was attributed to the higher affinity of kaolin to water as confirmed by the water contact angle of 22.8° compared to commercial PES UF membranes (contact angle = 65.5°). The modified membranes also exhibited less flux decline with surface water samples (lake and river water) and three synthetic organic matter solutions namely BSA, HA and SA compared to the unmodified membrane. Kaolin modified membrane also showed almost the same rejection level for the organic substances in all tested samples. This highlights the potency of kaolin modified as an attractive alternative for current commercial membranes applications (Hubadillah et al., 2020). For oily water treatment applications, Hubadillah et al. (2020) synthesized a low-
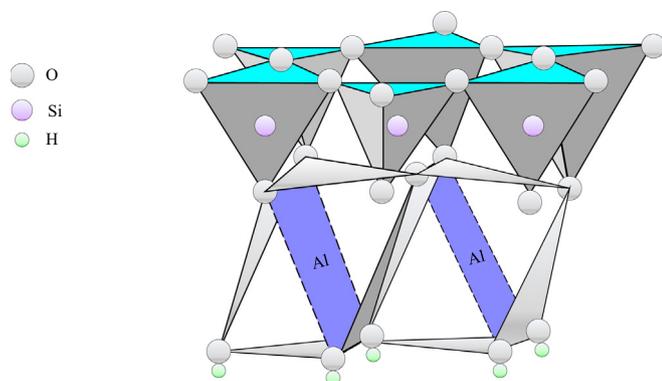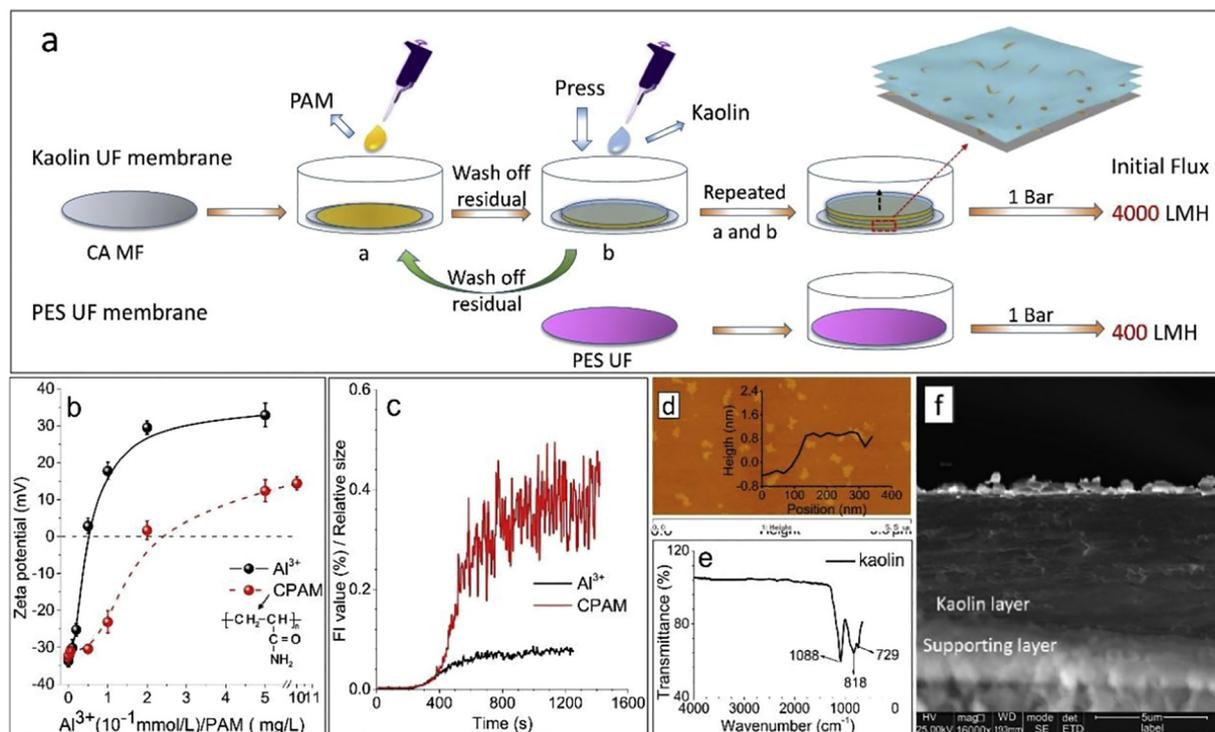
Fig. 4. 2 D kaolin membrane synthesis and characterization as reported in (Liu et al., 2019): (a) preparation steps; (b) zeta potential with two cross-linking agents (c) flocculation index of the two cross-linking agents; (d) Atomic force microscopy (AFM) analysis (e) FTIR-ATR spectrum of kaolin; (f) Scanning electron microscopy images of the produced membrane.
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cost kaolin hollow fibre membrane using sintering and phase inversion methods. The modified membranes manifested higher water flux (320 LMH) and 99.99% removal efficiency for both TOC and turbidity as well as 91.8% of COD for oily wastewater samples. T. K et al. (2019) fabricated PES nanocomposite membranes by incorporating nano zero-valent kaolin (nZVI: Kaolin) and Fe^{+2} supported kaolin (Fe: Kaolin) into a PES ultrafiltration membrane. The PES nanocomposite membranes were used for the separation of metal ions (Ni^{+2}, Cu^{+2}, and Cd^{+2} ions solution). The results disclosed that the modified membrane's hydrophilicity increased. Adding nZVI- Kaolin and Fe-Kaolin at different loading (0.015 wt% and 0.03 wt %) in the PES dope solution formed large finger-like morphology at the top layer and a sponge-like structure at the bottom. Higher water flux was also observed for all the modified membranes with a stable magnitude after 5 h of operation using dead-end cell filtration. The flux of Ni^{+2} Cu^{+2} and Cd^{+2} metal ions synthetic solution was improved along with the rejection of these metals due to the enhanced surface hydrophilicity and morphological structures. Saranya et al. (2012) prepared mixed matrix PAN membranes (PAN MMMs) using nano kaolinite. The kaolinite nanoparticles were obtained from naturally available kaolin by an intercalation and exfoliation method. The hydrophilicity of the modified PAN/nano kaolinite MMMs was higher (58.7° at 7.5 wt%) than that of pristine PAN membranes (70.2°) due to the inclusion of hydrophilic groups rich nano kaolinite in the PAN dope solution. The enhancement of the water productivity and both rhodamine B and egg-albumin protein selectivity due to nano-kaolinite incorporation would definitely suggest the viability of these MMMs for effective industrial wastewater treatment. These cost-effective PAN/ nanokaolinite membranes could also be harnessed for other energy intensive operations such as gas separation and pervaporation.

2.4. Zeolites

Zeolites are naturally occurring crystals with a microporous structure. Zeolites possess uniform pores and channel sizes. They are mainly composed of aluminium Al^{+3}, silicon Si^{+4}, and oxygen in their framework structure $[(SiO_2)(AlO_2)_x]_M \cdot yH_2O$ with a pore size range of 0.3–1 μm (Andaç et al., 2005). The atoms of aluminium and silicon are tetrahedrally linked with each other through shared oxygen atoms (Malamis and Katsou, 2013). The net negative charge of zeolite comes from the substitution of silicon ions (Si^{-4}) with aluminium cations (Al^{+3}) in the crystal structure. Similar to bentonite, zeolite also maintains a neutral charge by counterbalancing the negative charge with cations such as Na^+ and K^+ present in the structure cavities (Deer et al., 2013; Malamis and Katsou, 2013). The attractive feature of these cations is that they are readily exchangeable which bestows zeolite with the ability to remove cations with health concerns such as heavy metals. The zeolite framework can neutralise the positive charges of protons by providing negatively charged defect sites, which eventually form Bronsted acid sites (Koekkoek et al., 2012). Besides, it encloses the cavities occupied by water molecules and large ions that can move allowing ion exchange (Malamis and Katsou, 2013). The framework structure can also give rise to some properties within the pores such as hydrophobic, hydrophilic, and basic or acidic nature. Thanks to these combined properties, zeolites can selectively adsorb materials based on their shape, geometry, and chemical properties (Ramsay and Kallus, 2000). These features have resulted in zeolites being used in various applications including construction, agriculture, catalysis in the processes of oil refining and fine chemical synthesis, ion exchangers, separation of heavy metals from wastewater, desalination process, absorbent for softening and water treatment, and medical applications (Ciobanu et al., 2008; Han et al., 2009; Mumpton, 1999; Urtiaga et al., 2003).

Owing to its excellent permeability, selectivity, strong mechanical structure, and high thermal stability, the incorporation of zeolite into membranes has attracted tremendous attention in the past few years (Liu et al., 1996; Ma et al., 2012; Sano et al., 1994). Zeolite binding with polymeric matrix mostly takes place with the aid of facilitating agents. For instance, Yong et al. (2001) added 2,4,6-triaminopyrimidine to zeolite and polyimide mixture as compatibilizer, while Muntha et al. (2018) used (3-aminopropyl)-trimethoxysilan to condition zeolite making it reactive with PSF by replacing hydroxyl group (-OH) with an amine group

($-$NH$_2$), see Fig. 5. In both cases, silicate is the active group in the zeolite structure that is normally involved in forming bonds with membrane structure. Also, the dominant interaction in the binding of zeolite with membranes' materials is hydrogen bonding forming between the externally added amine group with an oxygen-containing functional group in membranes' structure. Due to its abundant negatively charged sites, uniform pore size distribution, and channel structure, zeolite was a perfect choice for membrane modification applications (Sun et al., 2018).

Zeolite micro and nano particles were incorporated into polymeric matrices in three different methods, namely mixed matrix membranes (MMMs), composite fibres, and pore-filled membranes (Ahmadiannamini et al., 2017; Maghami and Abdelrasoul, 2018b). MMMs are considered to be an effective option for the water treatment industry as they possess characteristics of both polymeric membranes and inorganic fillers (He et al., 2022). Damayanti et al. (2016) fabricated zeolite nanofiltration membranes for laundry wastewater treatment. The zeolite-based nanofiltration membranes manifested a higher membrane performance for micropollutants removal. It is worth mentioning that only a limited number of studies investigated the application of zeolite MMMs for water treatment purposes. One important design aspect that needs to be considered when using zeolite as a filler is the necessity of reducing the clay particle size to match the thickness of the active layer of the member. In this way, the composite can benefit from zeolite nanochannels as an effective flow path for water (Madhumala et al., 2014; Pechar et al., 2006). Zeolite does not swell in water and can form a suspension that is easily used to prepare inorganic membranes (Dong et al., 2006). The attractive qualities of zeolite such as high absorptivity of cations, availability of nanochannels for effective water filterability, and ease of regeneration and fouling removal make it a valuable multifunctional additive for MMMs synthesis. Zeolites

Fig. 5. Zeolite binding sites with polymeric membranes through: (a) application of compatibilizer and (b) zeolite conditioning. The figure was constructed based on the information presented in (Muntha et al., 2018; Yong et al., 2001).

have also well-defined porous structures and offer the mobility of alkali and alkaline earth metals, to compensate for a net negative charge between silicon and aluminium ions in the framework. This indeed makes zeolites excellent adsorber for the removal of many target compounds (Liu et al., 2018).

The construction of membranes with natural zeolite involves two categories of prerequisite processes associated with obtaining the desired geometry (e.g., shaping and grinding) and structure of the final crystal (e.g., sintering). It is worth noting that most studies that examined the application of zeolite as an additive for polymeric membrane focused on gas separation application. However, this work focuses on the zeolite-polymer composite application for water treatment purposes. Roque-Malherbe et al. (2006) prepared multi-layer ceramic MF membranes by adding different particle sizes of natural ground zeolite. The commonly reported sintering temperature range for zeolites is 800–900 °C (Abdullayev et al., 2019). The literature reveals that the pore size of inorganic membranes prepared with zeolite ranges from 0.3 μm (Hristov et al., 2012) and 1.1 μm (Dong et al., 2006). The pore size of membranes can be enlarged using pore-forming agents such as starch. The use of pore-forming agents during the preparation of zeolite membranes is important as the zeolite porosity can be very small after sintering due to glass formation (Chandrasekhar and Pramada, 2001). Ghaee et al. (2017) used zeolite NPs with TFN to improve hydrophilicity and water permeability. Results showcased that the permeability of the produced membrane was nearly doubled by the addition of nano zeolite as compared to the pristine TFN. Another research conducted by Adam et al. (2018) showed that the prepared zeolite hollow fibre membranes have a robust mechanic reflected by high flexural strength in the range of 8.5–51 MPa depending on the sintering temperature. The higher the sintering temperature, the stronger the produced membrane was. This is a high flexural strength compared to membranes made of clays such as kaolin. A study conducted by Abdullayev and co-workers reported a flexural strength range of 15–63 MPa for kaolin membranes prepared at a sintering temperature of 1200–1500 °C (Abdullayev et al., 2019).

Table 3 presents an overview of studies that tested zeolite as an additive for membranes. Overall, zeolite addition was found to increase membrane hydrophilicity which is likely to be due to the ability of zeolite to absorb water molecules through hydrogen bonding (Yurekli, 2016). There is an exception where one study reported a slight increase in hydrophobicity (Ma et al., 2012). Zeolite addition was also found to improve membrane antifouling characteristics. This is associated with an increase in hydrophilicity that increases pollutants-water interfacial tension. The improvement in salt rejection upon the addition of zeolite is ascribed to the formation of the dense membrane surface and porous support layer. This may also sometimes affect the flux negatively. The way zeolite is added to polymer solution could also affect the final composite structure. It was found that adding zeolite to the organic phase influences the interfacial polymerization reaction and results in a leaf-like structure (Lind et al., 2009). Such a practice was found to improve the miscibility of the mixture and consequently led to the formation of this structure (Dong et al., 2015).

2.5. Arabic gum (AG)

Arabic Gum, also called Acacia gum (AG) is a hydrophilic natural, biodegradable, biocompatible, and eco-friendly material that is extracted from the gummy exudation produced by some acacia trees species, especially *acacia Senegal* (Ali et al., 2013; Idress et al., 2021). This hydrophilic polymer is also found in some species of leguminous trees growing in the Sudan and Sahelian regions (Ribeiro et al., 2019). AG essentially consists of polysaccharides ($\sim$97%) with high molecular weight and a proteinous fraction ($\sim$3%) (Islam et al., 1997). The primary mono-saccharides in AG include arabinopyranose (Ara), glucuronic acid (Glca), galactose (Gal), and rhamnose (Rha), and the primary amino acid types are alanine, arginine, glutamic acid, glycine and histidine (Islam et al., 1997). AG has a carbohydrate fraction that is hydrophilic and a protein fraction that possesses hydrophobic properties. Due to its amphiphilic properties, AG has received
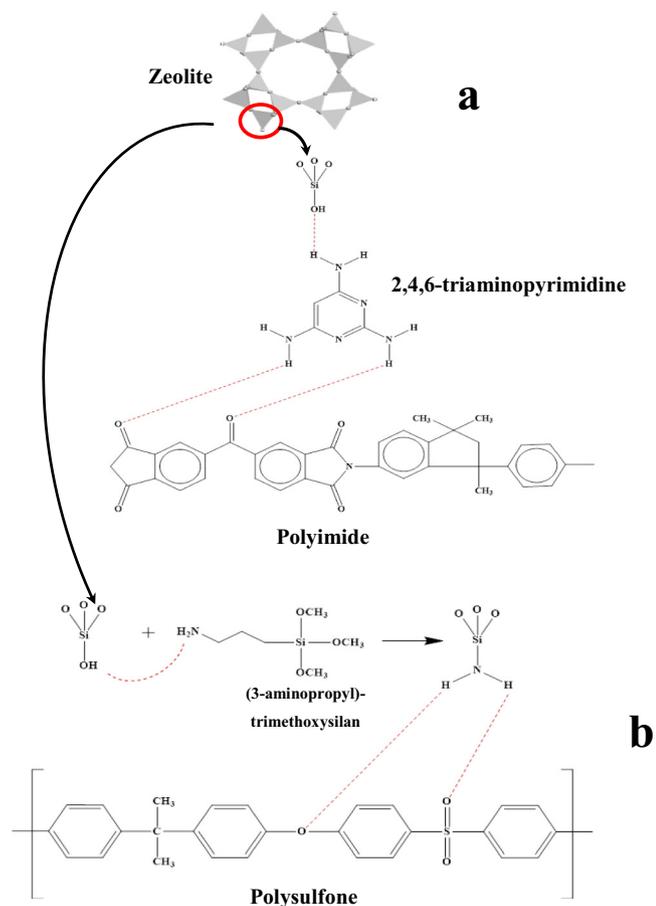
Table 3
Summary of research work conducted on zeolite incorporation into polymeric membranes.

Additives	Membrane materials	Feed	Improved membrane feature	Flux enhancement	Rejection enhancement	Ref
Porous UiO-66, Zeolite 4A, and UiO-66 and Zeolite 4A	Polysulfone mixed matrix membranes	Humic acid solution (HA)	Increased surface hydrophilicity, higher water flux, and rejection of HA, excellent anti-fouling properties.	119% increment as compared to pristine PSF	97% and 99% at 0.5 wt% and 1 wt% of UiO-66 and Zeolite4A	(Anjum et al., 2020)
SAPO-5 zeolite	Polyurethane membranes	Synthetic wastewater	Reduced membrane thickness and lower pore diameter in membrane active layer.	Higher flux: (7.41×10^{-4}) LMH as compared to pristine membranes (6.34×10^{-4}) LMH	–	(Ciobanu et al., 2008)
NaA zeolite nanoparticles	Polysulfone RO thin-film composite membranes (RO TFN)	2000 ppm aqueous solutions of polyethylene glycol (Mwt = 200 g/mol (PEG 200), 2000 ppm salt mixture of NaCl and MgSO$_4$	Reduced surface roughness, increased surface negative change, and hydrophilicity. The extent of the effect became more pronounced as the zeolite percentage increased.	Higher water permeation flux: $2.1 \pm 0.1 \times 10^{-12}$ m Pa^{-1} s^{-1} to $3.8 \pm 0.3 \times 10^{-12}$ m Pa^{-1} s^{-1} for TFN membranes at low and high contents of zeolite as compared to TFC membranes (2.1 ± 0.1)	A slight enhancement in salt rejection was achieved for the resultant RO membranes as compared to TFC Rejection for all the solutions is more than 90%	(Jeong et al., 2007)
NaA zeolite	poly (phthalazinone ether sulfone ketone) composite ultrafiltration (UF) ((PPESK)	Synthetic solutions of PEG and Titan Yellow dye (700 Mwt, 100 mg/L)	Improved antifouling properties, increased hydrophilicity, reduced thickness forming denser skin and porous sublayer.	340 to 246 LMH when the contents of NaA from 0 to 5 wt%	Increased rejection from 77.1% for unmodified membranes to 96.8% for composite membranes	(Han et al., 2009)
Zeolite nano-particles	Polysulfone	Metal solutions consist of Ni^{+2} and Pb^{+2}	Reduced porosity, increased pore size, increased hydrophilicity and wettability, and increased sorption capacity.	45 LMH/bar for PSf10–30 membrane as compared to pristine PSf (23.2) LMH/bar)	Increased Pb adsorption by 5 times and doubled Ni adsorption	(Yurekli, 2016)
NaY zeolite	Polyamide active layer: FO membranes	Salty solution 500 mg/L NaCl solution	Slight increase in membrane hydrophobicity and surface roughness especially at low zeolite loading.	Increased flux for zeolite loading up to 0.1 wt%/v by about 50% for both feed solution cases of deionised water and diluted NaCl solution.	Salt rejection was reduced by about 5% for the maximum zeolite loading of 0.4 wt%/v. Lower loading of zeolite resulted in further decrease in salt rejection.	(Ma et al., 2012)
NaY zeolite	RO membranes	Salty solution (2000 ppm NaCl solution	Enhanced hydrophilicity	Doubled water flux for zeolite loading of 0.15 wt%	Achieved salt rejection of 98.8% with zeolite loading of 0.15 wt%.	(Dong et al., 2015)

huge attention from many researchers recently (Bhushette and Annapure, 2017; Gashua et al., 2015; Sanchez et al., 2018; Shi et al., 2017).

AG has recently been used for enhancing membrane performance in applications such as gas separation, desalination, and other processes. Owing to its amphiphilic and anti-bacterial properties, AG has been used for improving mass transfer conditions in the region adjacent to the membrane surface (Suleiman et al., 2013). AG could bestow better surface hydrophilicity, improve porosity, and pore size, and improve anti-fouling properties of membranes while reducing membrane fouling and surface roughness. This

is due to the existence of the hydroxyl functional groups (-OH) that are responsible for the binding of AG with membrane structure, as illustrated in Fig. 6 (Idress et al., 2021). Sabri et al. (2019) fabricated a PSF membrane with different concentrations of AG using the phase inversion method. The results demonstrated that the flux increased from 50 LMH for pristine PSF to 130 LMH for modified PSF, and this was attributed to the hydrophilic nature of AG material. Besides, biofouling was inhibited on PSF/AG membrane surface due to the antibacterial effects of AG. In another research, waste PVC membranes were modified with the biopolymer/AG
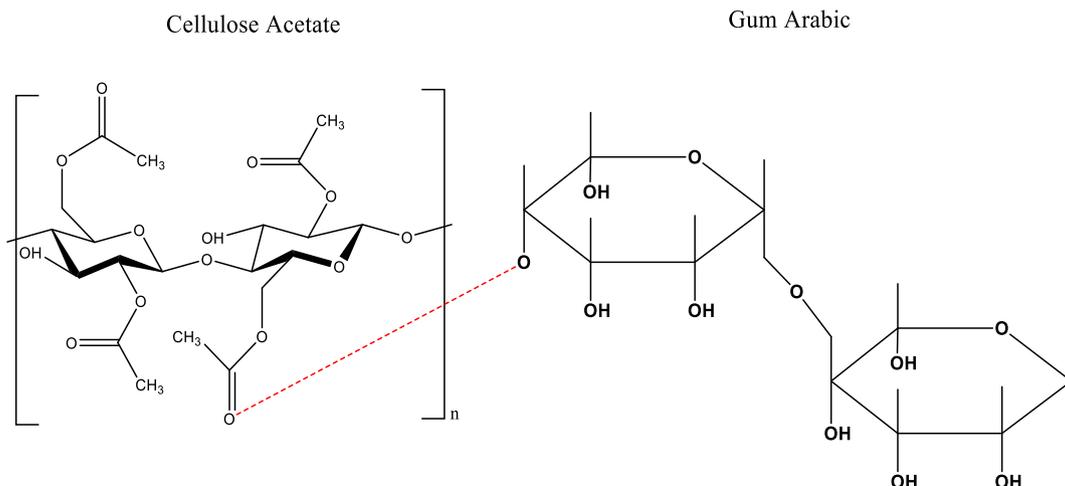
Cellulose Acetate   Gum Arabic

Fig. 6. Bonding site between Gum Arabic and cellulose acetate as a model membrane. The figure is constructed based on the information provided in (Idress et al., 2021).

for the removal of organic matter from water. The modified membrane exhibited a noticeable enhancement in flux from 51 LMH for neat PVC membranes to 98 LMH for modified PVC membranes due to the partial hydrophilic characteristic of AG (Aji et al., 2020). AG has also been applied as a pore-forming agent for synthesising polysulfone membranes (Manawi et al., 2017b). It was shown that using AG as a pore-forming agent did not only improve the porosity of the manufactured membranes, but also its hydrophilicity and consequently the permeability of the membranes.

A summary of the studies conducted using AG as a natural additive is given in Table 4. It is clear from the table that the addition of AG to membranes increases their hydrophilicity, structural strength, pure water flux, and salt rejection. In addition to this, AG possesses anti-bacterial properties and amphiphilic nature that help in reducing fouling in biological and organic forms. It was also reported that the addition of AG to polymeric membrane increase their mechanical stability. This can be explained by the abundance of the hydroxyl group in the AG structure that can participate in crosslinking the polymeric structure through hydrogen bonding (Falath et al., 2017). This behaviour of AG makes the produced composite less porous. However, AG behaviour and the final product properties depend on the polymer type used in the process and the casting process applied. For instance, when AG was used with PES and the phase inversion technique was applied, AG behaved like a non-solvent agent owing to its high affinity to water (Manawi et al., 2017a). This led to faster exchange and phased separation resulting in higher porosity.

2.6. Lignin

Lignin is an organic polymer that is widely available in nature (Li et al., 2021). Lignin forms an integral part of the plants' cell walls (Moo-Young, 2019). Lignin is naturally synthesized by plants and accumulates in the secondary walls of specific plant cells such as xylem vessels, fibres, and tracheids (Calvo-Flores et al., 2015). Lignin has an important role in maintaining the structural integrity of plants through fixing polymers in the wood plant cells which gives the wood its rigidity, serving as glue for holding cells together, maintaining hydrophobicity of cell walls, and acting as an antimicrobial agent that protects plants from biodegradation (Calvo-Flores et al., 2015). Lignin is isolated from wood by a combination of the physical and chemical processes represented by milling (ball or vibratory) and dioxane/water extraction follows by solvents purification (Caballero et al., 2003).

Table 4
Summary of recent studies on AG application as an additive for polymeric membranes.

Additives	Membrane materials	Water type	Improved membrane feature	Flux enhancement	Rejection enhancement	Ref
GO/AG	Polysulfone membranes (Psf)	Humic acid solution (HA)	AG addition alone results in a slight increase in membrane hydrophilicity, increased porosity, and pore size. Mixed with Go led to a further increase in the abovementioned characteristics.	Water flux increased by more than double with GA addition alone. Mixing with GO increased flux further.	HA rejection increased by about 4%. Mixing with GO deteriorated rejection further	(Chai et al., 2021)
GGA (Gum Arabic graphene)	PPSU membrane	Sodium Alginate	Increased surface roughness, decreased pore size, decreased hydrophilicity, and a slight improvement in thermal stability.	Best composite ration improved pure water and sodium alginate flux by ca. three times	Increased rejection by a maximum of 50% for 0.15 wt% of GAA. However, a further increase of GAA wt% to 0.25 decreased the rejection.	(Ali et al., 2021)
AG	PES	Synthetic solution consists of water and lead nitrate (500PPb)	Increased porosity, pore size (up to 0.5 wt% beyond which pore size decreased), increased hydrophilicity and negative surface charge, improved anti-biofouling properties, and increased mechanical strength. Permeate flux by 130%.	Increased flux by more than double for AG loading of 1% wt	Increased membrane rejection by more than double for AG loading of 3% wt.	(Manawi et al., 2017a)
AG	Cellulose acetate/vinyl triethoxysilane modified with graphene oxide	Pb (II) salt solutions	Increased thermal stability, porosity, hydrophilicity, and anti-biofouling properties. Reduced membrane compactness.	Flux increment by approximately 40% for the best GA loading percentage (8 wt%)	Increase of rejection by about 40% for best GA loading percentage.	(Idress et al., 2021)
Nanocrystalline cellulose (NCC)/AG	PVA RO membranes	boron solution (500 mg of boric acid/L)	Increased hydrophilicity, membrane stiffness, chlorine resistance, and anti-biofouling tendency. Reduced porosity in high loading percentages.	increased the flux from ca. 8.7 LMH to about 21 LMH for the best NCC/AG ratio of 1.3	Increased rejection by about 40% for the best NCC/AG ratio	(Asim et al., 2018)
AG	Polyvinylchloride (PVC)	humic acid (HA) (100 mg/L)	Improved thermal and mechanical stability. Increased hydrophilicity and reduced fouling propensity.	Increased flux by about 92% for a loading percentage of 3 wt%.	Increased rejection by about 15% for the highest AG loading percentage of 5 wt%.	(Aji et al., 2020)
AG	Polysulfone	20 ppm Congo red (CR)	Increased porosity, pore size, and hydrophilicity.	Increased water flux by almost double for membrane prepared with N-methyl-2-pyrrolidone (NMP), and by about 30% for membrane prepared with dimethyl sulfoxide (DMSO)	Increased rejection by about 2% for both solvents.	(Foong et al., 2021)
AG	Cross-linked PVA RO membranes		Decreased porosity, pore size, and surface roughness. Increased membrane stiffness and hydrophilicity. Improved biofouling and chlorine resistance.	Increased the flux by approximately 45% for AG loading percentage of 0.9 wt%.	Increased salt rejection by about 40% for AG loading percentage of 0.9 wt%.	(Falath et al., 2017)

Around 50 tons of industrial lignin is produced annually from biorefining and pulping industries (Li et al., 2021). However, most of the lignin is burned as cheap fuel or discharged directly as a waste product that contaminates the environment (Norgren and Edlund, 2014). If lignin is converted into a high-value material, it can be of great significance to mitigate environmental pollution and replace or supplement fossil fuels (Norgren and Edlund, 2014). Lignin possesses many outstanding properties including strong thermal stability, high carbon content, biodegradability, good stiffness, and good antioxidant activity (Albatrni et al., 2021). These properties have made lignin a good candidate to be used in different applications including fuels, agricultural, food packaging, and disposable products (Bottero et al., 2006; Li et al., 2019). It is also used in many forms of heat stabilizers, moulding stabilizers, light stabilizers, adsorbents, dispersants, antioxidants, surfactants, and other functional materials prepared from lignin (Kochkodan and Hilal, 2015; Ong et al., 2016; Rana and Matsuura, 2010).

Lignin is bio-renewable and environmentally friendly material that is suitable for polymer composites formation. In the composites polymer-lignin, lignin has some hydroxyl groups (-OH) attached to the aromatic ring, which provide great interactive sites with various kinds of polymers at the interface to attain better compatibility and obtain a more homogeneous composite membrane material (Chen et al., 2019; Kai et al., 2016; Thakur et al., 2014; Zhao et al., 2018). The typical lignin structure and the common binding pathways with polymeric membrane materials are illustrated in Fig. 7. The first pathway was explained by Chen et al. (2019) that a thermally induced ring-opening of polybenzoxazine facilitated the binding with lignin through the cross-linking reactions between phenol and phenoxy groups of lignin with reactive imine ions. The second pathway was reported by the same research group (Chen et al., 2021), but this time they tested the binding of PVA with lignin. The PVA is bound to lignin through hydrogen bonding with the -OH group. The crosslinking reaction was facilitated using maleic anhydride. The latter might have been used to increase the compatibility as lignin and PVA were found to form immiscible blends. The binding of cellulose with lignin was also reported to take place through the second pathway (Bilal et al., 2022).

The lignin composite membrane materials are not only cost-effective but also versatile. Composite lignin/SPEEK membranes have been shown to have a tight pore structure compared to conventional Nafion membranes while allowing enhanced proton transport (Ye et al., 2019). Alkaline lignin has been deposited onto the surface of the separative polyamide layer of the RO membrane via hydrogen bonding and π-π to simultaneously improve the water permeation flux, salt rejection, and anti-fouling properties of the RO membrane (Zhang et al., 2017b). Also, Lignin has been used as an interlayer spacer for engineering the nanochannels of GO membranes used for separating water from biofuels (Guan et al., 2018).

The recent attempts at lignin applications for membrane modification purposes are summarised in Table 5. The addition of lignin to membranes increases their hydrophilicity (Ding et al., 2016), resistance to solvents and oxidants (García et al., 2017), thermal resistance (Ding et al., 2016), porosity (Chen et al., 2021), and resistance to biofouling (Colburn et al., 2019). Most of these lignin traits are attributed to the functional groups available in its structure. For instance, the resistance to oxidants emanates from its ability to scavenge oxidants, such as radicals, through their interaction with the functional groups (Li et al., 2021). This has a positive impact on membrane resistance to chemical washing, which in turn could increase the membrane's lifespan. Similarly, the phenolic groups in the lignin structure possess antimicrobial properties that hinder the formation of biofouling (Colburn et al., 2019). Lignin has also been used as a surface functionalising agent for commercial membranes and was found to improve their fouling resistance. The lignin-modified polymeric membranes showed long-term stability toward salty solutions. The achieved results with lignin so far suggest that there could be promising research avenues open for investigation of membrane structure modification and surface functionalising of the commercial membrane. The main shortcoming of lignin is the high glass-transition temperature that limits its application with thermoplastic compounds. At high temperatures, the lignin structure turns brittle, and this would weaken the structure of the lignin-polymer composite (Collins et al., 2019). This issue can be rectified through mixing lignin with synthetic and natural polymers, using plasticizers, or through chemical modification (Collins et al., 2019).
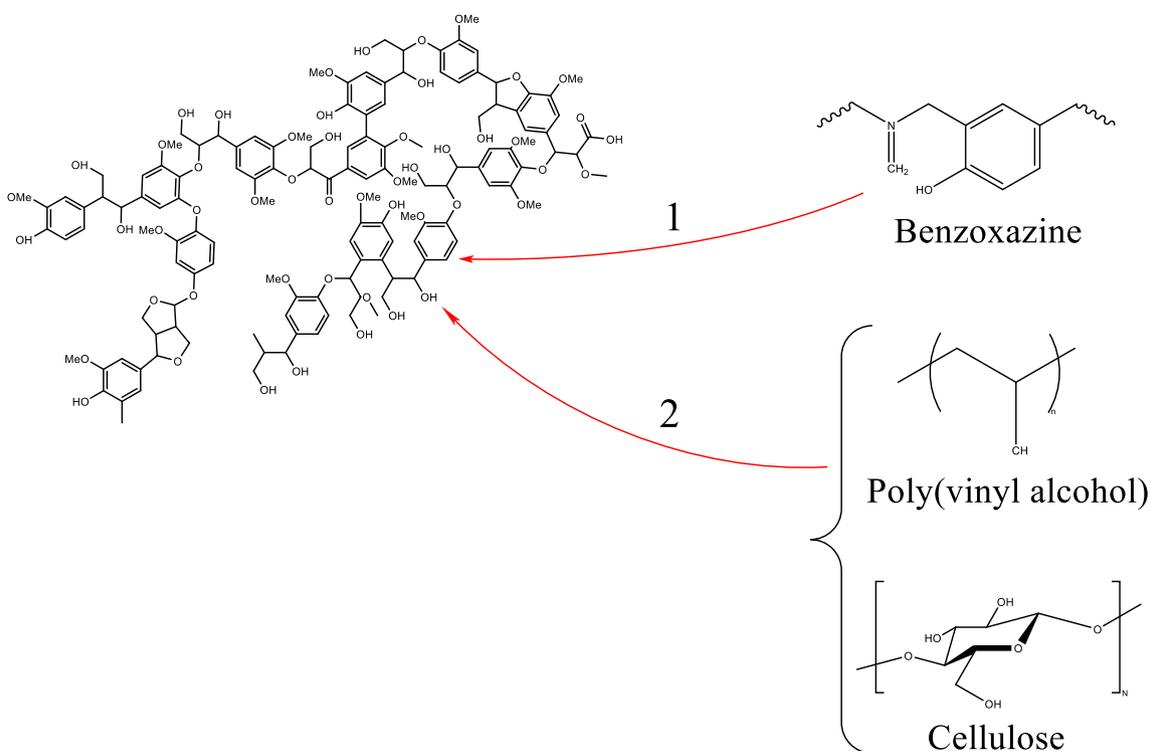

Fig. 7. Lignin structure and its common interaction pathways with membrane materials. The figure was constructed based on the information reported in (Chen et al., 2019; Chen et al., 2021).

Table 5
Overview of previous studies' outcomes for using lignin in membrane technology.

Additives	Membrane materials	Feed type	Improved membrane feature	Flux enhancement	Rejection enhancement	Ref
Lignin	PVA pervaporation desalination	Synthetic water contains salt (3.5 wt% NaCl)	Reduced porosity of support layer	18.5 LHM for 3.5 wt% NaCl solution. While for seawater the flux was 20.4 LMH	Almost 100% (99.95% of salt rejection) with concentration of 3.5 wt% NaCl. The same rejection was achieved for seawater	(Chen et al., 2021)
Lignin-cellulose nanofibrils (LCN)	PES UF membranes	BSA solution	Increased hydrophilicity, mechanical strength and maintain thermal stability.	Increase pure water flux by about 125% for lignin loading of 1.2 wt %.	Low lignin loading of up to 0.4 wt% did not affect BSA rejection, however increasing this to up to 1.2 wt% decreased the rejection by about 3%	(Ding et al., 2016)
Zeolite nanofibers	Lignin based membrane	Polystyrene microparticles in water	Adding zeolite increased membrane hydrophilicity, mechanical strength, and porosity.	With the addition of 1 wt% of zeolite, pure water permeability reached 38,259 ± 409 LMH/bar	About 8% without zeolite and approximately 95% with 1 wt% zeolite.	(Bahi et al., 2017)
Lignin	crosslinked polybenzoxazine membranes	Synthetic water contains tetrahydrofuran (THF)	Increased membrane hydrophilicity.	Permeation flux increased by 2.1-folds.	An increase of 3.4-folds in Pervaporation separation index	(Chen et al., 2019)
Lignin	crosslinked polybenzoxazine (PBz)	1 wt% water in toluene	Increased hydrophobicity and increased porosity led to weakening mechanical strength.	Achieved permeation of more than double what is reported in the literature for the same experimental settings and similar membrane material, but without lignin.	–	(Su et al., 2019)
Lignin sulfonate	cellulose membranes for preparing composite and commercial nanofiltration NF270 for coating	Humic acid (100 mg/L)	Improved biofouling resistance for cellulose membrane composite and reduced the negative charge when applied as surface functionalizing material for NF270.	Increased normalized flux by roughly 20–30% for composite membrane. Decreased water flux slightly when lignin sulfonate was applied as a surface functionalizing material	Improved the flux recovery with NF270 by significant mitigation of irreversible fouling.	(Colburn et al., 2019)

2.7. Aquaporin (AQP)

Aquaporins (AQPs), also defined as transmembrane water channel proteins, amphiphilic molecules, or a major of intrinsic proteins (MIPs), are a family of integral membranes that are ubiquitously present in the animal and plant kingdoms and allow water molecules to transport and reject of ions (Gonen and Walz, 2006; Verkman and Mitra, 2000). These channel proteins form pores in the membrane of biological cells, mainly facilitating the transport of water molecules between cells. AQP water channel proteins provide a passageway to more than 3×10^9 water molecules/s while maintaining high rejection of solutes in cell membranes (Borgnia et al., 1999; Fane et al., 2015). The two companies Aquaporin A/S and AquaZ companies founded in Denmark, represent the first manufacturers of AQP membranes on an industrial scale. AQP membranes polarized tremendous attention, particularly in Europe. This attention was translated into a large joint collaborative project funded by the European Commission (known as the MEMBAQ project). The project brought together several bodies including research institutes (DHI Water & Environment), universities from across Europe and Israel, and industry representatives (e.g., Anjou Recherche). They have investigated the incorporation of AQP into membranes for water and wastewater treatment and energy production applications (Habel et al., 2015). The project lasted for 3 years and a half from October 2006 to March 2010. Although the project resulted in 15 publications that deepened the understanding of AQPs membranes synthesis and performance, testing the produced membranes beyond the lab scale was not achieved during the period of the project (MEMBAQ HP, 2022). 0.09pt? > It was reported in the literature that the development of AquaZ coincided with the same period. AquaZ company was established in 2006 based on the patent filed by Carlo Montemagno in which he delineates the concept of integrating AQPs into polymeric membrane matrix to produce a new generation of membranes known as biomimetic membranes (Montemagno et al., 2004.). While Aquaporin A/S company continued to grow and currently is producing a variety of large-scale solutions for water treatment, AquaZ seems to struggle to maintain its presence in the market. A recent web search for AquaZ has returned results showing that AquaZ was on the Gust platform in 2018 to attract investors after the exit of Danfoss Innovation in 2010 (Habel et al., 2015).

Different types of AQPs are used in producing membranes, particularly in biomimetic membranes to form aquaporin biomimetic membranes and they showed attractive results. AQP embedded membranes can be divided into two types (Fig. 8): (1) AQP integrated into the active layer of a membrane known as the supported membrane layer (SML), in which the ultra-
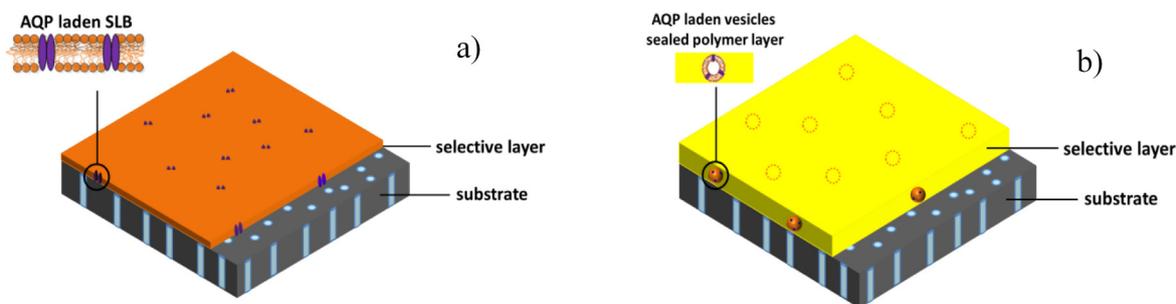

Fig. 8. Schematic diagram of AQP membrane configurations a) AQP supported lipid bilayers (SLBs) embedded in a thin active membrane layer and b) AQP vesicle immobilized within a dense membrane active layer (Tang et al., 2015) with copyright permission from Elsevier.

Table 6
Literature overview on AQPs applications for membrane modification.

Additives	Membrane materials	Water type	Improved membrane feature	Flux enhancement	Rejection enhancement	Ref
Aquaporin Z (AQPZ)	PAN NF	Synthetic MgCl$_2$ solution (200 ppm), glutathione separation at different pH (4–9)	Increased membrane surface roughness due to partial embedment of liposomes.	6.13 LMH in comparison with control LBL membranes (3.28) LMH, 60% improvement in water flux without using liposomes in the presence of AQPZ.	>95% of MgCl$_2$, 93% for glutathione rejection.	(Sun et al., 2013c)
Aquaporin Z (AqpZ) proteins and 1,2-dioleoyl-sn-glycero-3-phosphocholine (DOPC) (AqpZ-DOPC	NF-270 membrane	Salty solution consists of water and NaCl (1 mM)	The produced AQP membrane had similar surface roughness as NF after applying filtration at 1 bar due to the infusion of the lipids on the NF surface.	3.6 LMH/bar	20% NaCl rejection	(Li et al., 2012)
AqpZ-ABA	Cellulose acetate	Salty water consists of water and NaCl	Integration of AQP vesicles induced elemental change in the membrane structure indicated by an increase in nitrogen content.	34.2 LMH/bar	32.9% NaCl rejection	(Zhong et al., 2012)
AqpZ-ABA	AQP polymer on alumina	200 ppm NaCl	Integration of AQP vesicles induced elemental change in the membrane structure indicated by an increase in nitrogen content.	8.2 LMH/bar	45.1% NaCl rejection	(Duong et al., 2012)
AqpZ-DOPC/DOTAP	A mixture of poly (ethylenimine) (PEI), poly(sodium 4-styrene sulfonate) (PSS), and polyacrylonitrile (H-PAN)	Salty water (NaCl, 500 mg/L)	Increase of phosphorous and nitrogen content in the membrane structure. Decrease of zeta potential. Presence of unruptured vesicles, which may increase the surface roughness.	5.5 LMH/bar	75% NaCl rejection at 4 bars	(Wang et al., 2015)
AqpZ-DOPC	PS	Salty water (NaCl, 10 mM)	Increased surface roughness.	4 LMH/bar	96% NaCl rejection at pressure 5 bar and 98% at 10 bar pressure	(Zhao et al., 2012)
AqpZ-DOPC	Polydopamine (PDA) layered on PAN support substrate	Salty water (200 mg/L) and MgCl$_2$ solution (200 mg/L)	Reduction in PAN porosity due to PDA coating and increase in membrane surface roughness upon addition of AqpZ vesicles.	A significant drop in the permeability from 455 LMH/bar for PDA/PAN to 9.1 LMH/bar after the incorporation of AqpZ with AqpZ/lipid of 0.1.	66.2% NaCl rejection and 88.1% MgCl$_2$ at 5 bar pressure	(Sun et al., 2013b)
AqpZ-POPC/ POPG/cholesterol	A mixture of polyanion polymers (PAH and PAA) with poly cation polymer (PSS) on the PAN support layer	MgCl$_2$ solution (200 mg/L)	Increase in membrane surface roughness.	About 6 LMH/bar, an increase of 60% in the flux due to the incorporation of AqpZ	Rejection of ~96% of MgCl$_2$	(Sun et al., 2013c)
AqpZ-DOPC	poly(amide–imde) (PAI) and PEI	MgCl$_2$ solution (1000 mg/L)	Thermally stable membrane tested at a temperature of 70 °C for 2 h.	AqpZ membrane had slightly lower flux compared to PAI/PEI (32.0.7 LMH vs. 30.5 LMH)	AqpZ membrane achieved higher salt rejection than PAI/PEI (93.8% vs. 95.6%)	(Li et al., 2014)
AqpZ	Amine functionalized cellulose acetate (CA)	Two solutions (NaCl and MgCl$_2$ (200 mg/L)	High mechanical strength and stability during filtration tests.	Membrane with vesicle incorporation had pure water permeability of 14.3 LMH/bar, whereas the integration of AqpZ increased the flux to 22.9 LMH/bar	The respective NaCl and MgCl$_2$ rejection of vesicle imprinted membrane increased from 28% and 42%, to 39% and 51% after the addition of AqpZ.	(Xie et al., 2013)
AqpZ-ABA	polycarbonate with gold coating FO membrane	Salty solution 6000 mg/L NaCl feed	Increasing membrane roughness.	Increasing flux for both FO and PRO as the draw solution concentration increased to approximately 35 LMH and 45 LMH for, respectively as the draw solute concentration increased from 0.4 to 0.8 M	Salt reverse flux (g/m^2.h) increased for FO and PRO from 1.4 and 5.2 to 3.5 and 8.3, respectively when solute concentration increased from 0.4 M to 0.7 M.	(Wang et al., 2013)
AqpZ-ABA	Polycarbonate tracked-etched FO	Feed solution of 200 mg/L of NaCl and 0.3	Slight decrease in membrane roughness.	Water flux increased as the AqpZ:ABA decreased	Rejection of NaCl increased from 98.8% to	(Wang et al., 2012)

(continued on next page)

Table 6 (*continued*)

Additives	Membrane materials	Water type	Improved membrane feature	Flux enhancement	Rejection enhancement	Ref
	membranes	M of sucrose was used as draw solution		from 1:400 to 1:100. Water flux of AqpZ:ABA increased from ≈ 40 LMH to ≈ 140 LMH when salt concentration increased from 0.5 M to 2 M	99.1% when AqpZ:ABA increased from 1:100 to 1:400.	
AqpZ-POPC/ POPG/Cholesterol	Layered membranes of functionalized PAN and mixture of PSS and PAA	MgCl$_2$ as draw solution with various cocncentrations.	Enhance deposition of AqpZ onto membrane surface with the aid of magnetic field application.	Water flux increased as AqpZ percentage increased. The flux also increased from ≈ 40 LMH to 85 LMH when the draw solution concentration was raised from 0.25 to 1.5 M for AqpZ content of 2%.	Reverse salt flux increased as the percentage of AqpZ increased and reached eth maximum at 2% AqpZ. The reverse salt flus for the membrane with 2% AqpZ tripled when draw solution concentration increased from 0.25 to 1.5 M.	(Sun et al., 2013a)

thin bilayer structure enables the fast transport of water molecules through the AQP protein membranes, producing higher water flux and good biocompatibility with the membranes (Kaufman et al., 2010; Kaufman et al., 2014; Li et al., 2012). It is estimated that a biomimetic membrane composed of a lipid/aquaporin molar ratio of 2000:1 will have a water flux of 960 LMH, which is far superior to the FO polymer membranes. Although it is potentially doable to make large-scale industrial production, it poses low mechanical stability, scaling up difficulties along with the brittleness structure of the ultrathin selective layer. Also, there are difficulties to control the formation of defects during the formation process of the bilayer structure which are major challenges limiting their application in the membranes industry (Hansen et al., 2009a; Hansen et al., 2009b). (2) AQP vesicle enfolded membranes (VEMs), on the other hand where AQP vesicles (proteoliposomes or proteo-polymersomes) are encapsulated in a thick polymeric layer (Li et al., 2014; Sun et al., 2013b). This layer makes the membranes mechanically robust, minimising the formation of defects and facilitating the potential scalability of large-scale industrial production, and protecting the AQPs protein membranes from biological and chemical attacks (Wang et al., 2013; Zhao et al., 2012). However, the membrane water flux is sacrificed to a certain extent depending on the property of the protective layer matrix and other factors (Zhao et al., 2013). Additionally, the chemicals applied to immobilise the AQP vesicles may significantly impact the functionality of implanted vesicles (Tang et al., 2015).

Since AQPs first inception in the 1990s, their astonishing water transport capabilities and higher solute selectivity have attracted membrane scientists. The outstanding properties of AQPs such as scalability, and cost-effectiveness made them a great candidate for a variety of applications including membrane technology (Tang et al., 2015). In membrane manufacturing, AQP has been used potentially for the construction of next-generation membranes that must reject 100% of different types of solutes, except water, and are used precisely for desalination, water reuse, water purification (e.g. RO, NF, and FO) and pharmaceutical applications (Kumar et al., 2007). Kumar et al. (2007) demonstrated that AQPs integrated into polymer display higher water permeability flux (up to 2-fold) and higher solute rejection of approximately 100% as compared to the commercial RO membranes. However, despite all these features, some challenges are limited to their broad applications. One of these challenges is aquaporin protein suitability for membranes ultrathin membranes on a nanometre scale. Such membrane cannot withstand high pressure and harsh environmental conditions without the need for a support layer (Tang et al., 2015).

For aquaporin membranes to compete with commercially available membranes, they should ideally have superior water permeability that could adequately cut down on the energy cost of water treatment (e.g. achieving a lower energy requirement than the desalination figure of 2.6 kWh/m^3) (MacHarg et al., 2008). Having a high water permeability

would reduce the need for the number of filtration modules required for a certain capacity, and this in turn would reduce energy associated with other processes elements such as pumps and pipes operation and maintenance (Sun et al., 2013c). Recently, Cohen-Tanugi and co-workers conducted a modeling study to evaluate the effect of water permeability on energy savings for seawater RO (SWRO) and brackish water RO (BWRO) systems with respective recovery ratios of 42% and 65% and capacities of 300 m^3.day^{-1} and 140 m^3.day^{-1} (Cohen-Tanugi et al., 2014). They found that increasing water permeability three-fold could reduce the energy requirement by 15% and 46% for SWRO and BWRO, respectively. This signifies the importance of developing membranes with high water permeability such as AQP membranes. For more in-depth information regarding biomimetic membranes, readers are referred to these dedicated literature review reports on this topic (Abaie et al., 2021; Beratto-Ramos et al., 2021; Goel et al., 2021; Perry et al., 2015; Shen et al., 2014; Tang et al., 2015; Tang et al., 2013). Table 6 presents an overview of the studies reported on employing AQP in membrane technology.

2.8. Other natural additives

Other natural additives have been recently utilized for membrane performance improvement purposes such as chlorophyll. Chlorophyll is a green pigment that is available in plants. One of the common applications for chlorophyll is its medicinal use. The known sources of chlorophyll used for medicinal purposes are algae, alfalfa, and silkworm droppings. In membranes, it has been used as pore-forming agents to get porous membranes and improve the membrane structure. Recently, Azhar et al. (2021) fabricated mixed polysulfone matrix membranes by incorporating the extracted chlorophyll from spinach and PEG to enhance the PSF pore structure. The results of this study showed that the pore structure of PSF mixed matrix membranes was enhanced when the concentration of chlorophyll was 0.25 wt%. This also improved hydrophilicity and surface roughness. As a consequence, the water flux and rejection of humic acid increased considerably (230 LMH and 86% at 0.25 wt% chlorophyll in comparison with 175LMH and 80% for pristine PSF membranes).

3. Recycled additives

3.1. Biochar

One of the most attractive recycled materials that have significant potential in water treatment applications is biochar. Biochar is a green porous material of carbonaceous structure that is formed by the thermochemical disintegration of naturally abundant biomass in a zero or low oxygen environment (Lehmann and Joseph, 2009; Shackley et al., 2012; Verheijen et al., 2010; Xiang et al., 2020). A variety of biomass sources can be

employed for biochar production such as wood, manure, algae, crop and forest residues, sewage sludges, and organic municipal solid waste (Colantoni et al., 2016; Xiong et al., 2019). The methods used for biochar preparation include pyrolysis, hydrothermal carbonization, gasification, microwave heating, and torrefaction (Fang et al., 2018; González et al., 2017; Mohan et al., 2014).

Biochar has polarized remarkable research attention due to two reasons: (1) Biochar is a low-cost material, and environmentally friendly adsorbent owing to its high surface area, and porosity, copious active surface functional groups, simple production process, and wide distribution-specific compositions (Cha et al., 2016; Inyang et al., 2016; Pan et al., 2021), and (2) biochar can store carbon in a stable form that can offset greenhouse gas emission (Creamer and Gao, 2016; Xiang et al., 2020). The properties of biochar (physical and chemical) are affected by many factors including biomass feedstock, type of thermochemical decomposition, temperature, and duration. Furthermore, biochar properties can also be affected by the applied pre-treatment processes of the raw materials and post-treatment of the produced char (Wang et al., 2017; Xiang et al., 2020; Yu et al., 2019). Due to its economic benefits, biochar has been used in different applications including; (1) soil amendment, (2) mitigation of climate change, (3) production of alternative energy to fossil fuels (4) waste management, (5) as a catalyst and absorbent agent for water and wastewater treatment (Ghaffar et al., 2015; Ghaffar et al., 2018; Lehmann and Joseph, 2009).

Recently, biochar has been utilized for improving membrane performance. Ghaffar et al. (2018) developed free-standing PVDF/biochar composite membranes in uniform loading and controlled compositions where the phase inversion technique was applied to prepare the composite membranes. The wood biochar was pyrolysed at temperatures of 300 °C and 700 °C (denoted as B300 and B700, respectively), and then mixed with PVDF membranes at different concentrations (10%, 30%, and 50%). Results showed that the biochar particles are uniformly distributed throughout the membrane surface and across its thickness. The resultant membranes had a porous structure and higher mechanical strength. The membrane pure water flux increased with increasing the amounts of biochar. For B300, as the biochar: PVDF mass ratio increased from 1:9 to 5:5, the flux increased from 4825 to 5411 LMH. The same increase in the biochar: PVDF ratio for B700 resulted in flux enhancement from 5823 to 6895 LMH. It is worth mentioning that the pure water flux of the pristine membrane was 4634 LMH. B300 membranes were more hydrophilic and possess higher surface energy compared to B700 membranes (58.84–60.31 m.J/m^2 vs. 56.32–51.91 m.J/m^2). The biochar membranes showed outstanding adsorptivity toward RhB dye with an adsorption capacity of 47–187 mg/g. He et al. (2017) developed a hollow fibre biochar/PSF mixed matrix membrane. Micro-sized bagasse biochar was synthesized first and then incorporated into PSF membranes to form mixed matrix membranes. The resulting membranes were more hydrophilic as compared to pristine PSF membranes while a higher permeation flux was achieved. The adsorption of lead and copper for all the mixed matrix membranes increased as the pH increased with achieving the maximum adsorption capacity at pH >4.5. The adsorption equilibrium was achieved at 7 and 12 h for copper and lead, respectively. The presence of humic acid minimally affected adsorption, while the ionic strength adversely affected the removal. Additionally, the feed concentration and cross flowrate significantly affected the removal efficiency in a continuous filtration mode. The increase in flow rate and feed concentration resulted in a reduction in the volume of treated permeate that had concentrations of copper/lead below the regulated levels for drinking water. In a recent study, Mohammadi et al. (2021) developed an adsorptive mixed matrix polymeric membrane by introducing biochar (made from *Rosmarinus officinalis* leaves (BRM)) into a polymeric membrane (PVC-BRM). The resultant membranes were used for phosphorus separation and compared with ZnO as reference. The maximum adsorption capacity of BRM was 78.24 mg/g, and the adsorption of phosphate was endothermic and followed Langmuir and Freundlich isotherms. This indicated that multiple physical and chemical mechanisms were involved in the adsorption. Both BRM and PVC-BRM membranes showed high removal efficiencies of

phosphate in single and multi-component matrices. The phosphate adsorption was pH-dependent and improved in acidic conditions. The adsorption capacity had a direct correlation with an initial concentration of phosphate for BRM and PVC-BRM, although removal efficiency decreased. Desorption efficiency was low in both BRM and PVC-BRM. However, 75% regeneration was achieved after washing BRM with NaOH solution. There is an interesting approach reported by Huang and co-workers on synthesising biochar/geopolymer membrane through in-situ simultaneous carbonization of both materials (Huang et al., 2020). Although what they produced was a very small-scale membrane and only tested in dispersion experiments, the approach has a great potential to prepare cost-effectively biochar mixed matrix inorganic membranes. The exciting thing about this idea is using biochar as a catalyst to facilitate the degradation of recalcitrant contaminants such as tetracycline with aid of oxidants (e.g., H_2O_2). The challenge with this approach is the need for a long retention time to achieve high removal which can be through a combination of adsorption and oxidation mechanisms. In this case, maybe the use of a multi-stage unit with a high concentration of oxidants can offer a solution.

More research about the integration of biochar into membranes is provided in Table 7. Unmodified biochar increases the hydrophilicity of membranes. The increase of hydrophilicity upon biochar incorporation is attributed to the availability of hydrophilic functional groups such as -OH and -COOH in biochar structure (Gu et al., 2021). However, modified biochar with 3-Aminopropyltriethoxysilane (KH-550, silane coupling agent) decreased the hydrophilicity (Lan and Wang, 2018). The increase in thermal stability upon biochar addition is attributed to the inorganic content of biochar that does not degrade even at high temperatures (Lan et al., 2016). Biochar was also found to increase surface roughness when added at high loadings. In this case, biochar particles agglomerate at the active layer (Gu et al., 2021). The agglomeration of biochar on the active layer may increase the risk of biofouling formation as biochar porous structure offers a good environment for microbial growth.

3.2. Fly ash (FA)

Fly ash (FA) is one of the most abundant waste materials that is generated as a by-product of the firing of coal in power plants (Joo Kim et al., 2014). It is a heterogeneous material that is mainly composed of Al_2O_3, SiO_2, CaO, and Fe_2O_3 (Ilic et al., 2003). The precise structure of FA can vary significantly depending on the coal it is generated from (Joo Kim et al., 2014). Globally, about 80 million tons of FA are produced and only a small amount of the produced FA is utilized in construction applications such as cement alternatives manufacturing, pavement and concrete, and brick manufacturing (de la Rocha et al., 2021; Rawlings et al., 2006). The silica-rich compositions of easily available FA are a valuable source for porous additives that can be applied for membrane modification (Abdullayev et al., 2019). Due to its outstanding properties such as sphericity (1–100 μm as shown in Fig. 9), non-toxicity, porosity, lightweight property, and high strength, FA is regarded as a cost-effective material for many applications including the removal of dyes and heavy metals from wastewater, and adsorption of volatile organic compounds from the air (Kim et al., 2013; Sultana et al., 2012; Visa et al., 2010). If fly ash is not handled appropriately, it can pose serious health risks and harm the ecosphere (Sears and Zierold, 2017). Thus, there is a need to manage this waste to decrease its environmental impact (Dong et al., 2009).

Some researchers have used FA as a cost-effective replacement for common adsorbents such as granular activated carbon (Ahmaruzzaman, 2009; Lindgren and Norbäck, 2002). While others find this waste material is a great candidate for ceramic membranes fabrication (Barbieri et al., 1999; Ilic et al., 2003). The nature of FA with the abundance of metals oxides afford attractive opportunities for low-cost inorganic membrane fabrication for wastewater treatment applications (Abdullayev et al., 2019). The structures made from FA have a pore size distribution that is suitable for the functionality of microfiltration membranes and support layers. Nevertheless, most of the work done on FA utilization in membrane technology was directed toward synthesising support layers owing to its strong

Table 7
Overview of up-to-date studies on using Biochar in different membrane applications.

Additives	Membrane type	Water type	Improved membrane features	Flux enhancement	Rejection enhancement	Ref
Ball-milled biochar (BMBC)	TFC membrane consists of PDA/BMBC	100 mg/L of tetracycline hydrochloride (TCH), oxytetracycline (OTC), Chlortetracycline Hydrochloride (CTC) and ciprofloxacin hydrochloride (CFH) was used as feed solutions,	Increased hydrophilicity and roughness of the membrane. The flux of TCH, OTC, CTC, and CFH improved upon BMBC incorporation. The extent of improvement increased with increasing BMBC loading.	Pure water flux increased with increasing the filler loading reaching a maximum of double that of the pristine membrane.	Rejection of TCH, OTC, CTC, and CFH slightly increased with integrating BMBC up to a loading concentration of 4 mg/mL.	(Zhang et al., 2021)
Tree bark biochar (BB) grafted with $NH_2(CH_2)_3Si(OC_2H_5)_3$ (KH-550)	polydimethylsiloxane (PDMS) membrane	Synthetic solution consists of Ethanol/water	Increased hydrophobicity (many due to the grafting materials) and thermal stability.	Increased the flux by about 40%	The separation factor increased by a maximum of 50%. This was achieved with a filler loading of 3%.	(Lan et al., 2016)
Biochar prepared from lodgepole pine bark modified with KH-550 and $CH_2 = CH-Si(OCH_3)_3$ (YDH-171)	PDMS membranes	Synthetic solution consists of Ethanol/water	Increased hydrophobicity (many due to the grafting materials) and swelling degree. YDH-171 had a slightly higher effect compared to KH-550.	Flux increased with increasing filler loading concentration to a maximum of ca. 40% for both grafting materials.	The separation factor increased by a maximum of 50% and 67% for biochar modified with KH-550 and YDH-171, respectively. This was achieved with a filler loading of 3%.	(Lan and Wang, 2018)
Biochar/Kevlar	nano-BC/Kevlar nanofiltration	Saline solution contains NaCl and Na_2SO_4, and a mixture of dye solution methyl blue, reactive blue, and 19Congo red.	Increased surface roughness and hydrophilicity of membrane.	Deionised water flux increased by about 13 times with the highest biochar loading of 10 wt%. BC loading of 5% that had the best salt and dye rejection had the same flux as that of Kevlar.	highest salts and dyes rejection improvement was achieved with biochar loading of 5%. The increase in all dyes' rejection was about 5%. The rejection of NaCl and Na_2SO_4 increased by approximately 36% and 12%, respectively.	(Gu et al., 2021)
Biochar-SiO_2 core-shell particle (BCNPs)	PDMS membranes	Synthetic water consists of ethanol/water	The addition of BCNPs has led to remarkably improvement in PV performance of composite membranes. When a BCNPs content was 5 wt% for a 10 wt% ethanol solution at 40 °C, the best PV performances gained were the separation factor of 11.9 and the corresponding permeation flux of 227 $g \cdot m^{-2} \cdot h^{-1}$ (0.227 LMH)	227 LMH		(Lan et al., 2017)
Micro-scale biochar	PS hollow fibre membrane	Simulated waste water containing dye (methylene blue) (MB)	The MMM achieved high removal of MB under pH range of 4–10. An excellent reusability for removal of MB.	295.75 L/m²	the final desorption rate can reach above 90%, which is 10% higher than that by single organic desorption solution (data not shown)	(He et al., 2018)

mechanical structure. The pore size of membranes fabricated from FA ranges between 0.18 μm and 7.28 μm (Abdullayev et al., 2019). Jedidi et al. (2009) prepared MF membranes using fly ash for treating textile industry wastewater. A microporous support layer was prepared first then an active MF layer of FA was made by the slip casting process and coated on the support layer. Both the active and support layers were made from the 4 wt% fly ash, 30 wt% PVP as a binder, and 66 wt% of water. The membrane permeability was 475 LMH/bar. The obtained permeate quality for the two membranes was almost the same with the COD and colour removal of 75% and 90%, respectively. Fang et al. (2011) prepared a free-cracking tubular supported ceramic microfiltration from FA with methylcellulose and water using a slip-casting method. The results showed that three key factors such as withdrawal speed, contact time, and slip concentration influenced the properties of the prepared membrane. Applying several coating whey syntysising membranes may reduce the pore size. For instance, in the study conducted by Fang et al. (2011) showed that the average pore size of a single-coated FA membrane was 0.90 μm with a permeation flux was of 1.70×10^4 LMH/bar, while a double-coated membrane had an average pore size of 0.77 μm with permeation flux of 1.56×10^4 LMH/bar.

Fly ash can be used to manufacture nanofiltration membranes when it is mixed with zeolite. Two research studies were conducted by Zhu et al. (2018) and Dong et al. (2009) have combined fly ash with zeolite, producing membranes with different pore sizes ranging from 0.93 μm to 2.2 μm for the former and 0.18 μm to 0.26 μm for the latter. Table 8 presents a summary of studies that used fly ash as a structural material for membrane fabrication. From the reviewed literature on the application of FA, it seems that its incorporation increases the hydrophilicity of the modified membranes which remarkably improves the membrane performance and endows the membranes with excellent thermal and mechanical properties.
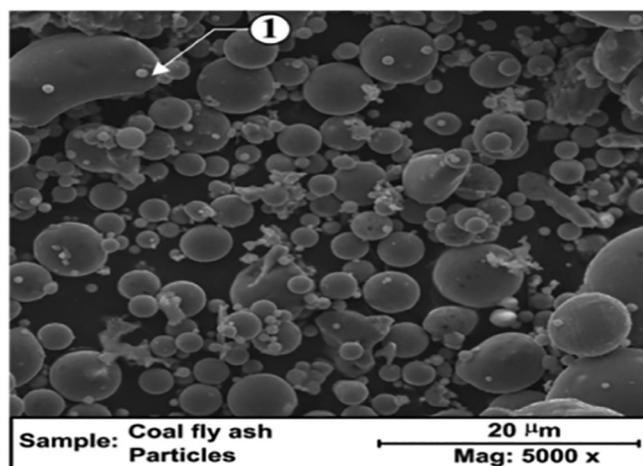
Fig. 9. SEM images of Fly ash with spherical structure (Golewski, 2017) with copyright permission from Elsevier.

4. Recycled waste as the main structural membrane materials

Although the focus of this work is on discussing the use of natural and recycled additives for improving membrane separation and structural properties, addressing the possible applications of recycled waste for producing skeletal membrane materials is equally important. Hence, we have presented here several recycled waste options that have been harnessed for producing membrane materials to provoke ideas and taking lead from these examples to develop this research area further.

4.1. Recycled polystyrene (RPS)

Polystyrene is a polymer that is generally used in civil engineering/architecture in the form of foamed polystyrene as Styrofoam for building isolation (Adamczak et al., 2020). Styrofoam is expanded polystyrene (EPS) and consists of 2% of polymer and 98% air. This product is mostly used in insulations and for manufacturing heat-insulating containers for food and packaging during transport (Adamczak et al., 2020). Polystyrene is also used in a large number of applications such as packaging for fragile goods such as electronics and the production of disposable products foam cups (Vakhshouri and Nejadi, 2018). All these products generate about 6200 kt of EPS in 2015, only 40% was used and the rest was wasted. Therefore, it is important to look for ways of reducing the impact of this waste on the environment by recycling them for other applications.

Recently, foamed polystyrene (FPS) has been used as the main material for the production of polymeric membranes for water purification. Various non-waste polymers like PSF, PES, PAN, PVDF, and others are used for the production of polymer membranes. The use of polystyrene is almost negligible. Rare studies have been published on using foamed polystyrene as basic material for manufacturing polymeric membranes. Ramos-Olmos et al. (2008) fabricated membranes using FPS as polymer source, *l*-methyl-2-pyrrolidone (NMP) as a solvent, and PEG and dioctyl phthalate as additives. The phase inversion technique was applied to prepare the RPS membrane. The resulting membranes have higher water flux after the addition of PEG with high molecular weights, as compared to the pristine polystyrene. The presence of additives affected the surface properties of the modified membranes. Adamczak et al. (2020) prepared ultrafiltration UF membranes consisting of RPS and single-walled carbon nanotubes modified with hydroxyl groups (SWCNT-OH). The prepared UF membranes were used to treat river surface water. The results showed that a slight increase in water flux was obtained for the RPS membrane due to its higher hydrophilicity. In another study, Ke et al. (2016) developed hydrophobic electrospun polystyrene nanofibrous membranes for desalination using the DCMD process. The results demonstrated the capacity to desalinate successfully seawater with a rejection rate of >99.99%. The membrane water flux decreased with increasing the membrane thickness and decrement of pore size under the chosen DCMD conditions for all feed solutions. In another research work, Huan et al. (2015) prepared electrospun polystyrene fibres by dissolving PS beads in a mixture of DMF and THF. The SEM examination of the produced fibres illustrated the existence of combined fibres in a strand-like structure that was indicated to be the reason behind the observed strong tensile strength of 1.5 MPa. It was also noticed that increasing THF: DMF resulted in stronger smoother fibres with no bulges, and this was ascribed to the formation of a more viscous polymer solution with the increase of these solvents ratio. The other synthesis aspect studied in this work was the effect of the electro-spinning voltage. It was noticed that increasing the voltage from 12 to 20 kV increased the hydrophobicity of the produced mat. Although Huan et al. (2015) study did not test the produced fibre mats for water treatment, they provided valuable results that can be of great benefit for preparing RPS membranes. These studies highlight the potency of PS waste as a cheap and environmentally friendly source for polymeric membrane fabrication.

4.2. Polyethylene terephthalate (PET)

PET is one of the widely used commercial polymers in various industries (Arahman et al., 2017). PET has been used in various applications due to its chemical stability in acids and organic solvents, remarkable mechanical properties, high transparency, and good gas-barrier resistance which

Table 8
Summary of previous studies' observations for integrating fly ash into membranes' materials.

Additives	Membrane materials	Water type	Improved membrane feature	Flux enhancement	Rejection enhancement	Ref
Fly ash (FA) and TiO_2	polyurethane nanocomposite membrane (PU)	$HgCl_2$ (10 ppm) and Pb $(NO_3)_2$ (5 ppm) in water; aqueous methylene blue dye (MB) (10 ppm)	Higher surface hydrophilicity, and excellent thermal properties. Higher membrane performance was obtained after the incorporation of TiO_2 and Fly ash.	At 5 psig, 1000 LMH for PU/FA as compared to pristine PU	–	(Kim et al., 2014)
coal fly ash (CFA)	Ceramic alumina membrane	Commercial corn-starch (0.25 kg/m^3) and particle diameter (12 μm)	Observed linear shrinkage and deteriorated mechanical strength on one hand, and higher porosity accompanied by higher water flux on the other hand.	116 for pristine membrane (CFA0) and 370 for modified membranes (CFA40)	99% of corn-starch solution	(de la Rocha et al., 2021)
fly ash particles	Crosslinked Polyethylene composite membranes	ethanol-water mixture	Good dispersion of fly ash into the membrane matrix. The highest water flux was achieved.	873 LMH	Separation factor (15.93)	(Kumar et al., 2020)
Fly Ash	Crosslinked polyamide membranes	Phenol solution	Good water flux and good chemical stability. The prepared membranes can efficiently separate phenol from aqueous solution	0.184 LMH/kPa(18.4 LMH/bar)	The removal efficiency of phenol was 92.3%	(Gupta and Anandkumar, 2019)

made it the most usually used material for the fabrication of organic membranes using electrospinning and track etching. The plastic bottles are composed of PET which can potentially be converted to membrane materials (Ding et al., 2012). Such repurposing of this waste does not only help to decrease the waste of plastic bottles but also reduces polymers' consumption. Recycling PET bottles for membrane production could significantly drop the cost of membranes production (Arahman et al., 2017). PET is a versatile material used in different forms such as electro-spun fibres fabrication as a nonwoven support layer (Passos de Oliveira et al., 2018; Strain et al., 2015; Zander et al., 2016), ion tracked-etched membranes (Espinoza et al., 2014; Zhang et al., 2016), support or coating layers for hollow fibre membranes (Hao et al., 2016; Liu et al., 2009; Zhou et al., 2018; Zhou et al., 2017), and currently in track-UV membranes (Wang et al., 2018). The characterization of PET membranes can be improved further through post-treatment such as the case in these studies where silver ions were added to the membrane fibre to install anti-bacterial properties in the membrane (Lin et al., 2011a; Lin et al., 2011b).

The utilization of recycled PET for membrane synthesis has been explored by a few studies. Rajesh and Murthy (2014) found that PET membranes without modification with a have poor mechanical properties. However, the addition of PEG with higher molecular weights resulted in higher permeation flux and a stronger structure. Kusumocahyo et al. (2020) examined using recycled plastic PET to fabricate UF membranes through the phase inversion technique. Their results showed that lowering the polarity of the nonsolvent by using different kinds of nonsolvent (water/ethanol, water/n-butanol, and water/n-propanol) increased water permeation. The membrane also demonstrated higher water flux by increasing the concentrations and the molecular weight of the additives for the casting solution. UF experiment was done using BSA as foulant model and the membranes showed rejection up to 91%. In another attempt, Kusumocahyo et al. (2021) prepared UF membranes from recycled PET bottles and PEG (Mwt = 400 KDa) as an additive and water/ethanol as a coagulation non-solvent. PEG plays a vital role in determining the properties of the produced membrane. Increasing PEG concentration reduced pore size but increased porosity and hydrophilicity of the membrane. These changes brought about an increase in pure water flux and BSA solution rejection. PET recycled bottles were also used to prepare nanofiber MF membranes using electrospinning technique (Zander et al., 2016). The membrane was used for separating latex beads with sizes ranging from 30 to 2000 nm. It was found that the membrane was capable of separating 99% of the beads with only gravity filtration. Xu et al. (2020) prepared electrospun nanofiber membranes from recycled Coca-Cola bottles and modified them through fluorination. The prepared membranes were used for membrane distillation (MD). The results showed that the membranes were highly hydrophobic with a contact angle of ≥130°, having higher water flux (11–23 LMH and higher rejection, almost 100%) compared to pristine membranes (10 LMH and 95% respectively). Pulido et al. (2019) prepared porous UF membranes from recycled PET for the filtration of PEG/water and PEG/DMF at high temperatures and with harsh chemicals. The PET membranes exhibited excellent resistance toward acidic and oxidative

agents in the presence of solvents such as DMF at a high temperature of 100 °C. This reflects the potency of recycled PET membrane in applications where high temperature and corrosive materials are present. UF membranes have also been fabricated using a recycled PET via TIPS method (Arahman et al., 2017). PVP was applied in 5 wt% in some of the prepared membranes to test its effect on the quality of the produced membranes. The dope solution conditions also varied, with and without evaporation. The addition of PVP produced a sponge-like structure in the membrane sublayer while casting without evaporation resulted in a more porous membrane. The membrane without PVP addition and evaporation had the highest flux of 97 LMH, whereas the membrane with 5 wt% PVP and evaporation resulted in the highest HA rejection of 76%. This highlights the potency of recycled PET membranes for surface water treatment; however, further refining of the synthesis and modification processes is still required to produce high-performing membranes. Kiani et al. (2021) have prepared nanofiltration membranes using PET/XA with two non-solvent solutions: methanol and water. The membranes were applied for the diltiazem removal from aqueous solution. The addition of 0.25–0.75 wt% of XA to the casting solution of membranes prepared in water and methanol increased porosity, thickness, and hydrophilicity of the membranes. Further increase of XA to 1 wt% decreased these properties. Adding XA made the membrane structurally weaker as indicated by tensile and elongation tests. Membranes prepared with water and methanol with XA of 0.75 wt% had the highest steady flux of ≈ 38 and 42 LMH, respectively. Diltiazem rejection was the highest (98%) with PET membrane with 0.25 wt% XA prepared in methanol, while the highest rejection (92%) for membrane prepared in water was achieved with 1 wt% XA. This is another example showcasing the importance of fine tuning the synthesis process of recycled PET membranes. PET is another plastic waste material that can successfully be used for fabricating membranes with the need of some modifications for improving the resultant membrane structure.

5. Natural and recycled additives versus common additives

Previous sections presented detailed discussion concerning the opportunities of harnessing natural and recycled waste materials as fillers and main materials for synthesising membranes for water treatment purposes. Here, we attempt to establish a comparison between the reviewed additives and their counterparts of common additives such as TiO_2, ZnO and Ag, $Mg(OH)_2$, SiO_2, CNTs (Jian et al., 2021), SWCNTs, MWCNTs, GO, rGO (Yang et al., 2018), $CaCO_3$, Fe, FeO, MgO, MOFs (He et al., 2022). The comparison is based on four criteria namely performance, energy cost, and impact on the environment and human health as illustrated in Table 9.

The addition of common, natural, or waste-based additives into the membrane matrix has a similar positive impact on the properties of the final product. MMMs normally have remarkably higher permeation flux and salt rejection, higher mechanical strength, selectivity, thermal stability, hydrophilicity, and chemical stabilities compared to pristine membranes. The porous and adsorptive nature of some natural and waste-based additives may give them a competitive edge over common additives in dye

Table 9
General comparison between natural, waste-based, and commonly used chemical additives in membrane technology.

Important aspects for consideration	Natural additives	Recycled waste additives	Common additives chemical additives
Improved performance indicator	- Improved membrane flux - Antibacterial activities - Better adsorption - Higher hydrophilicity - Mitigate membrane fouling Enhanced dyes and heavy metals removal	- Improved solute rejection - Enhanced removal of heavy metals and micropollutants - Improved membrane flux - Increased hydrophilicity when unmodified fillers applied - Increased separation factor in pervaporation	- Improved membrane flux - Higher hydrophilicity - Higher anti-fouling property - Improved solute rejection - Improve thermal and mechanical stability
Cost	Low cost of unmodified materials.	Low cost for unmodified and modified materials.	High cost
Environmental impact	Benign effects given they are extracted from naturally occurring materials.	Have a positive environmental impact given they are repurposed waste.	Production of these additives has a high global warming impact.
Health concerns	There are no reported health concerns associated with these additives.	May have little impact on health depending on the waste. BC made of agricultural waste has no health concerns.	Potential health concerns (e.g. TiO_2 Np).

removal and pervaporation applications. The strong catalytic and anti-bacterial effects and the large surface to volume ratio of nanoparticles such as TiO_2, Ag and ZnO (Basile et al., 2015) might make them a more attractive option than natural and waste-based additives for tackling biofouling and simultaneous removal and destruction of recalcitrant pollutants. In order to make a fair performance assessment of natural and waste-based additives as opposed to common additives, a direct comparison studies using the same composite mixture and treatment conditions are required. Such studies have not been present in the literature yet, but there are few attempts that compared inorganic membranes made of clays and other materials such as TiO_2 for water treatment applications. For instance, Suresh and Pugazhenthi (2017) reported that TiO_2 membrane had a better oil removal from oil wastewater compared to ceramic membrane prepared from a mixture of clays, quartz and calcium carbonate. They reported oil removal of 99% with TiO_2 in different pressure and cross flow velocities, while the oil removal of ceramic membrane ranged between 93% and 96% (increased as pressure and cross flow velocity increased). In this example study, TiO_2 appeared superior to clays. However, more studies are required to gain a better insight into how the different classes of additives compare against one another. The authors believe that different applications and environmental conditions would favour different additives and there also exists the possibility of applying hybrid composites where recycled or natural additives are combined with common additives.

Discussing the cost and energy requirements of additives can be a controversial topic as it involves many intertwine elements. However, it is important to shed light on the cost of only the additives without discussing the cost of processing, storage, handling, and disposal for the sake of establishing a comparison between the different additives categories. Common additives are too costly, and the processing of nanoparticles requires complex steps with poor reproducibility. These nanoparticles usually increase the manufacturing cost and there still exists a deliberation on whether the expense of these nanoparticles overshadows their advantages. For instance, the production cost of TiO_2 nanoparticles was quoted to be around 0.03–1.21 $/g (Kamali et al., 2021). This cost figure is relatively high in comparison to the cost figures of natural and recycled additives that will be discussed later in this section. It is noteworthy that the manufacturing cost of MMMs with some natural fillers is also high such as the case with zeolite (Maghami and Abdelrasoul, 2018a). However, the high cost of MMMs with zeolite can be lowered by utilizing simulation tools such as molecular dynamics simulation that helps in predicting the diffusive behaviours of the composite membrane. Natural additives are not expensive and easy to obtain due to their natural abundant availability. A low-cost range of 4–130 USD/m^2 was estimated manufacturing membranes from natural clays. For instance, the estimated cost of some natural clays such as kaolin and pyrophyllite is 4 USD/m^2, while the cost of other natural minerals such as quartz and sodium metasilicate is estimated to be 130 USD/m^2 (Vinoth Kumar et al., 2015). The cost of bentonite was also reported to be as low as 0.005–0.04 USD/kg (Shamsudin et al., 2019; Zaitan et al., 2008). The cost of recycled additives can be much lower than that of common and natural additives. For example, Rosales et al. (2017) reported biochar production costs as low as 0.076 $/kg. Even the modified biochar cost is still lower than that of common additives. The production cost of amine-modified biochar was reported to be 1.6 $/kg (Kamali et al., 2021).

The environmental side effects of common additives such as nanoparticles are another challenge that impacts their applications and has not been systematically or fully concluded. A thorough overview of the environmental impact associated with materials produced from different industries has become achievable through the use of the developing LCA tools and their growing inventories. A recent study conducted by Temizel-Sekeryan and Hicks (2020) on evaluating the global warming impact of silver nanoparticles (Ag Np) production showed that common production techniques have high CO_2 emissions of 500–1500 kg CO_2-eq/kg of Ag Np. In comparison, biochar production using different feedstocks and various portable reactors produced a negative global warming impact of −1000 to −2000 kg CO_2-eq/t of biochar (Sahoo et al., 2021). Recycled waste additives have far better environmental effects than common additives. This is one of the

attractive points that should encourage further research in harnessing such additives for membrane separation improvement.

The discharge of nanoparticles from the membrane into the treated water during filtration may affect human health and contaminate the environment. For instance, unmodified graphene, graphene oxide, reduced graphene oxide, carbon nanotubes, silver nanomaterials have shown great potential in different membrane applications (Chen et al., 2016; Farahani and Vatanpour, 2019; Sonawane et al., 2017); however, the environmental safety uncertainty due to present cytotoxicity to human and animal cells could reduce their broad range of applications. This has been confirmed by many research studies that proved these nanoparticles are genotoxic.

The effect of additives on human health is an important aspect that needs to be considered when comparing certain classes of additives to others. There is a risk of additives leaching into treated water which make them in direct contact with consumers. Some common additives such as AgNP and MOFs have been found to have toxic effects. It was reported that AgNP could accumulate in several body organs of tested animals such as kidneys, liver, lung, spleen, and brain (Rosales et al., 2017; Yu et al., 2013) reported that AgNP could accumulate in several body organs of tested animals such as kidneys, liver, lung, spleen, and brain. This could potentially lead to neurotoxicity, lung inflammation, and organs malfunction. Similarly, MOFs (e.g. nanoZIF-8) have been reported to have toxic effects on human cells (HeLa) (Sajid, 2016). Recycled additives such as biochar on the other hand were found to have no toxic effect on epigenetic earthworm and human cells (Amaro et al., 2016; Yang et al., 2019).

6. Conclusions and future directions

The application of natural and waste-based materials as additives and raw materials for membrane technology has been thoroughly discussed in this work. The focus has been directed toward water treatment applications. Though the topic is relatively new, especially for recycled waste materials, there is a decent number of attempts in the literature that proved its worth for expansion and development. The reviewed studies in the literature showed that natural and waste materials can be used in synthesising MMMs with minimal modifications, however, this may be at the expense of the final product quality. Sometimes the modification is necessary for improving the compatibility between fillers and membrane materials. Oxygen-containing functional groups are the main elements that bind different fillers with membrane materials. Hence, in most cases, modification of either fillers or membrane materials involves introducing these groups or other active groups such as amine groups. One important common observation in all studies is that performance of the produced MMMs depends largely on applying the appropriate fillers load that strikes the right balance between flux and rejection. The reported changes in membrane characteristics with natural and waste-based fillers are similar to those observed in common chemical additives (MOFs, TiO_2, AgNP, etc.). However, when comparing materials' cost, environmental impact, or effects on human health, natural and waste-based fillers outperform common additives.

The findings of the investigations reported in this work indicate that the application of natural and waste-based fillers is a nascent research area that deserves the attention of both research and industry. Several research directions should be adopted to develop this area. Establishing a universal database for estimating the cost of natural and waste-based materials is imperative to realistically evaluate the feasibility of utilizing such materials in membrane manufacturing and modification. Some materials such as biochar witnessed decent good progress with regards to this aspect, but still, there is a big variation in the reported production prices from different sources. Information concerning MMMs production cost on a large scale is also lacking. This may require support from main players in the membrane manufacturing industry to at least help in building tools for estimating production costs. A comprehensive cost analysis for fillers that encompasses not only extraction/manufacturing cost but also storage, handling, disposal, and other elements would have a great benefit in the advancement of this research field. A unified approach to studying the

environmental impact of different fillers should be followed by utilizing LCA tools. This would help both researchers and industry in making decisions for choosing materials with minimal environmental impacts. Studying the fate and impact of natural and recycled additives on human health and their effective exposure pathways (ingestion, inhalation, and dermal) is an important future research goal. In addition to the above, side by side comparative studies between common additives and natural and waste-based additives would be useful to have a better insight into how these groups of additives differ from one another in terms of advantages and shortcomings. Long-term testing of all additives with different conditions will also help to evaluate their stability and tolerance to real-life applications. Studying the compatibility of unmodified natural and waste-based additives with the elements of the active and supportive layer of membranes may generate useful information for possible ways of reducing the cost of membranes synthesis with such additives. The suitability of different casting techniques with natural and waste-based additives should also be studied to appropriate manufacturing conditions that result in the best compatibility between the additives and the continuous phase. These research directions need to be fulfilled to formulate a conclusive assessment of the potency of these fillers.

CRediT authorship contribution statement

Muayad al-Shaeli: Conceptualization, Methodology, Writing - Original and revised Drafts, **Raed A. Al-Juboori:** Conceptualization, Methodology, Visualization, Writing - Review & Editing. **Saif Al Aani:** Methodology, Writing - Review & Editing, **Bradley P. Ladewig:** Methodology, Writing - Review & Editing, **Nidal Hilal:** Methodology, Writing - Review & Editing.

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.

Acknowledgement

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