



## Review

# A comprehensive review on the synthesis and applications of ion exchange membranes

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## ABSTRACT

Ion exchange membranes (IEMs) are undergoing prosperous development in recent years. More than 30,000 papers which are indexed by Science Citation Index Expanded (SCIE) have been published on IEMs during the past twenty years (2001–2020). Especially, more than 3000 papers are published in the year of 2020, revealing researchers' great interest in this area. This paper firstly reviews the different types (e.g., cation exchange membrane, anion exchange membrane, proton exchange membrane, bipolar membrane) and electrochemical properties (e.g., permselectivity, electrical resistance/ionic conductivity) of IEMs and the corresponding working principles, followed by membrane synthesis methods, including the common solution casting method. Especially, as a promising future direction, green synthesis is critically discussed. IEMs are extensively applied in various applications, which can be generalized into two big categories, where the water-based category mainly includes electrodialysis, diffusion dialysis and membrane capacitive deionization, while the energy-based category mainly includes reverse electrodialysis, fuel cells, redox flow battery and electrolysis for hydrogen production. These applications are comprehensively discussed in this paper. This review may open new possibilities for the future development of IEMs.

## 1. Introduction

Ion exchange membranes (IEMs) are undergoing prosperous development in recent years. IEMs are broadly applied in various industrial applications. A desirable IEM is expected to possess high conductivity/low resistance, high ion exchange capacity (IEC), high permselectivity, high dimensional stability/low membrane swelling and water uptake, as well as high chemical, mechanical and thermal properties (Hosseini et al., 2017a; Jiang and Ladewig, 2017; Son et al., 2017). However, it is very challenging to prepare IEMs which have all the above merits. Various synthesis methods are developed aiming to meet some or all of the above requirements. On the other hand, the diverse applications of IEMs necessitate the development of different IEMs with desired properties.

As shown in Fig. 1, over 30,000 papers which are indexed by Science

Citation Index Expanded (SCIE) have been published on IEMs during the past twenty years (more details are provided in the supporting information). Especially, more than 3000 papers are published in 2020 alone, which discloses researchers' great interest in IEMs. Meanwhile, about 7% of the papers published in 2020 are review papers. The top-cited review papers in 2020 are summarized in Table 1. However, although many reviews papers have been published on IEMs, the number of review papers which extensively focus on the applications of IEMs are very limited. Given the diverse applications of IEMs, it is necessary and important to give an up-to-date review, which is beneficial to researchers and their research on IEMs. This paper aims to provide a review on the recent advances of IEMs. To be specific, conventional and special types of IEMs and their electrochemical properties and working principles are discussed, followed by membrane synthesis with particular focus on green synthesis, and finally, a comprehensive and critical

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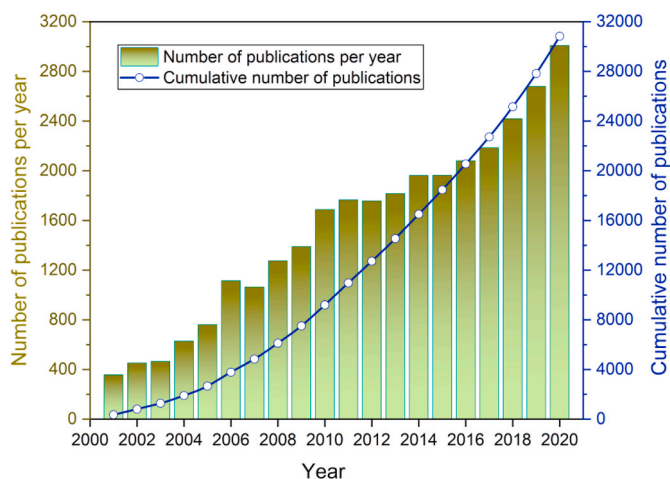


Fig. 1. Number of publications on ion exchange membranes during the past 20 years (2001–2020) (Jiang et al., 2018).

Table 1

Summary of the top-cited review papers published in 2020.

Reference	Summary	Research areas
Al-Amshawee et al. (2020)	This paper reviewed the process, principles, and setups of electro dialysis (ED) technology.	ED
Du et al. (2021)	This paper reviewed the degradation of catalysts in fuel cell and discussed materials-based solutions.	Fuel cell
Song et al. (2020)	This paper reviewed the bipolar plate materials and their preparation methods, flow field layouts, and application in proton exchange membrane fuel cell (PEMFC).	PEMFC
Falcão and Pinto (2020)	This paper reviewed the published models of proton exchange membrane (PEM) electrolyzers used to predict cell voltage, including reversible voltage, activation losses, ohmic losses and mass transport losses.	PEMFC
Shabani et al. (2020)	This paper reviewed the cost and energy recovery of PEMs used in microbial fuel cell (MFC) application.	MFC
Wang et al. (2020)	This paper reviewed the materials, designs, fundamental principles, and manufacturing processes of PEMFC.	PEMFC
Besha et al. (2020)	This paper reviewed the negative effects of multivalent ions on electricity generation of reverse electro dialysis (RED) and discussed the possibility of using monovalent selective membranes to solve these issues.	RED
You et al. (2020)	This paper reviewed the synthesis methods of alkaline-stable anion exchange membranes (AEMs).	AEM
Miller et al. (2020)	This paper reviewed critical materials and operating conditions for hydrogen production from AEM-based water electrolysis.	Electrolysis
Gurreri et al. (2020)	This paper reviewed applications of ED in wastewater treatment.	ED

review on the applications of IEMs is given, which may shed new light on the future development of IEMs.

## 2. Types and working principles of IEMs

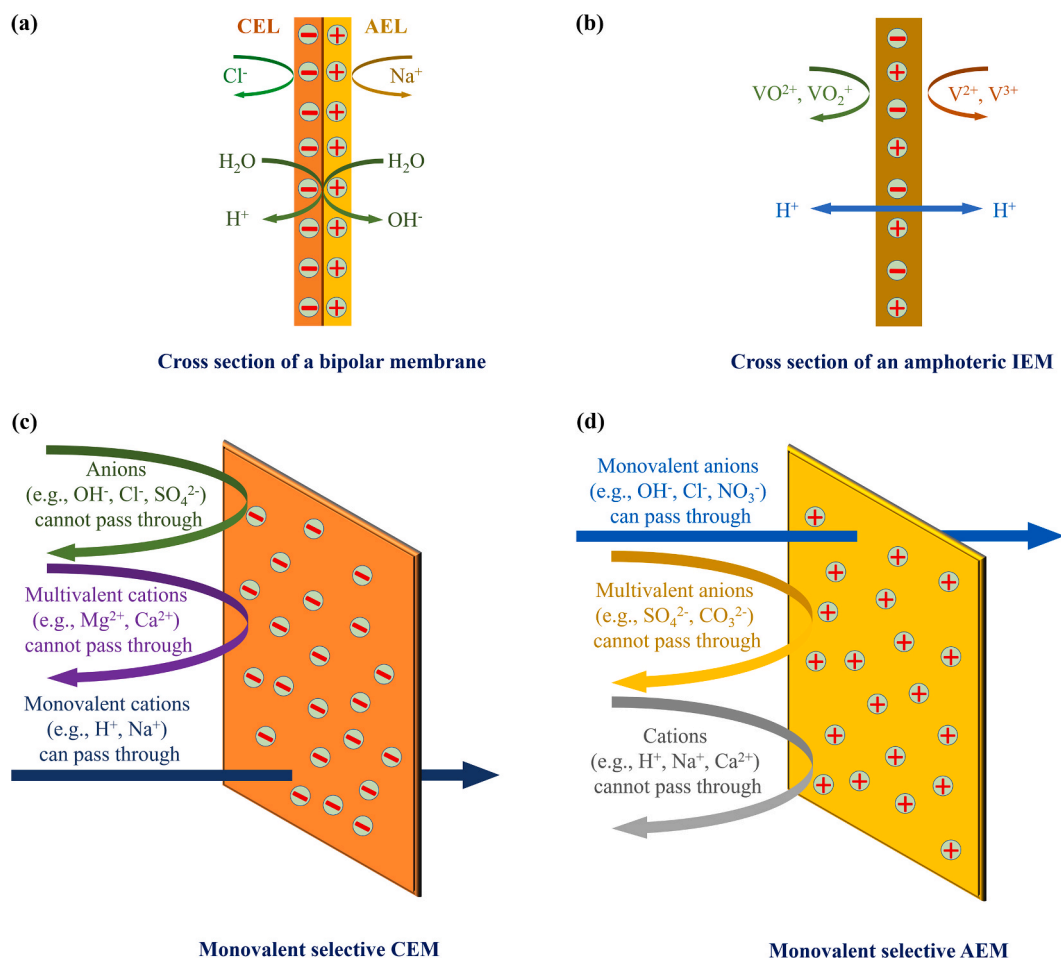
Based on the type of ions that is desired to pass through a membrane, there are two basic types of IEMs, i.e., cation exchange membranes (CEMs) and anion exchange membranes (AEMs). CEMs contain fixed anion groups and exchangeable cations in the polymers. In contrast,

AEMs contain fixed cation groups and exchangeable anions in the polymers. In addition to the common functional groups, there are some emerging types of functional groups for AEMs in recent years, such as metal-cation-based group (Hagesteijn et al., 2018). For example, Zha et al. (2012) synthesized positively-charged ruthenium complex ions that acted as the functional group for the AEMs, which demonstrated similar performance compared to conventional AEMs in terms of mechanical stability and anion conductivity. Li et al. (2020) prepared positively-charged zirconium-based metal organic frameworks that acted as the functional group for the AEMs, which showed great improvement in hydroxide conductivity.

In addition to common CEMs and AEMs, there are also a few special types of IEMs, including proton exchange membranes (PEMs), bipolar membranes, amphoteric IEMs (also known as charged mosaic membranes), monovalent selective IEMs, and mixed matrix membranes (MMMs). PEMs, as a common type of CEMs, are mainly used to conduct protons in fuel cells (Kim et al., 2015), which will be discussed in detail in the applications section. As its name indicates, bipolar membranes contain both cation exchange groups and anion exchange groups (Khoiruddin et al., 2017; Ramdin et al., 2019). As shown in Fig. 2, a bipolar membrane is composed of two layers, namely the cation exchange layer (CEL) which is similar to a CEM and anion exchange layer (AEL) which is similar to an AEM. When power is supplied, water will dissociate into hydrogen ions and hydroxide ions at the interface of the two layers (Pärnamäe et al., 2021a). As will be discussed below, bipolar membranes are mainly used for electro dialysis applications (Fu et al., 2014; Haddad et al., 2017). Similar to bipolar membranes, amphoteric IEMs also contain both cation exchange groups and anion exchange groups. However, as illustrated in Fig. 2, the cation exchange groups and anion exchange groups in amphoteric IEMs are “mixed” together, which is different with bipolar membranes where cation exchange groups and anion exchange groups are separated. Amphoteric IEMs are promising for vanadium redox flow battery applications (Liao et al., 2015; Nibel et al., 2017). Low permeability of vanadium ions and high proton conductivity are two key factors assuring membrane performance for vanadium redox flow battery applications. The main advantage of amphoteric IEMs over conventional CEMs lies in that amphoteric IEMs have lower permeability of vanadium ions (Wang et al., 2014). Also, compared to conventional AEMs, amphoteric IEMs have higher electrical conductivity (Hu et al., 2012). However, mechanical stability is also a very important factor, but it is still challenging to prepare amphoteric IEMs with excellent ion conductivity and mechanical stability simultaneously, and more efforts are needed to address this issue (Liu et al., 2021).

Another special type of IEMs is monovalent selective IEMs. As indicated by its name, monovalent selective IEMs have the ability to separate monovalent ions from solutions and retain multivalent ions in solutions (Nie et al., 2017; Radmanesh et al., 2019). As shown in Fig. 2, monovalent selective IEMs can be further classified into monovalent selective CEMs (Ge et al., 2014; Li et al., 2015) and monovalent selective AEMs (Güler et al., 2014; Zhang et al., 2018). Monovalent selective IEMs can be used to remove arsenic and nitrate ions from groundwater (Cohen et al., 2017), to concentrate reverse osmosis brines via electro dialysis (Xu et al., 2018b), to generate energy through reverse electro dialysis (Güler et al., 2014), etc.

In recent years, a novel type of IEMs is being developed, which incorporates nanofillers into the polymer matrix, and the resulting membranes are known as mixed matrix membranes (MMMs) (Hosseini et al., 2013; Ran et al., 2017). As shown in Table 2, various nanofillers are used to prepare MMMs. Compared to common IEMs, the incorporation of nanofillers in MMMs can tune membrane properties like surface hydrophilicity, improve membrane stability in harsh environments like high temperature, and correspondingly improve separation performance (Alabi et al., 2018). However, the amount of nanofillers must be carefully controlled which could otherwise decrease membrane mechanical strength and negatively affect electrochemical properties.



**Fig. 2.** Schematic illustration of some special types of IEMs. (a) Cross section of a bipolar membrane. (b) Cross section of an amphoteric IEM. (c) Monovalent selective CEM. (d) Monovalent selective AEM.

Common electrochemical properties of IEMs include electrical resistance/conductivity, permselectivity, IEC, current density, etc. Among these, electrical resistance/conductivity and permselectivity are two of the most important ones. Especially, permselectivity refers to the ability of an IEM to selectively transport counter-ions instead of co-ions (Avci et al., 2020a). To be specific, CEM allows cations to pass through but blocking anions and neutral molecules, while AEM allows anions to pass through but blocking cations and neutral molecules. The reason is that, CEM contains fixed functional groups which are negatively charged. Due to charge repulsion, anions are less likely to pass through a CEM. The permselectivity of AEM is similar. As mentioned earlier, high permselectivity and low electrical resistance are desirable. Table 3 summarized the most important electrochemical properties of some reported IEMs in literature and commercial IEMs (Güler et al., 2013; Hosseini et al., 2019, 2017a; Zhang et al., 2019).

Also, different IEMs with different properties are commercially available and there are several famous manufacturers of IEMs such as MEGA (Czech Republic), FumaTech GmbH (Germany), Tokuyama (Japan), etc. For example, FumaTech GmbH has a variety of IEMs available, where some are manufactured with no reinforcement while others are manufactured with reinforcement (e.g., polyester, polypropylene, PEEK and PTFE). Generally, reinforced membranes possess stronger mechanical strength but the membranes are also much thicker, and as a result, the electrical resistance is also higher compared to non-reinforced membrane. In applications such as membrane capacitive deionization (MCDI) where membrane mechanical strength is not a deciding factor for overall system performance, non-reinforced membranes are more favourable as it is thinner and has lower electrical

resistance.

As summarized in Table 4, the types of IEMs can also be categorized based on the operating conditions/environment. For example, as will be discussed later, operating fuel cells at high temperature can improve the overall performance, and therefore it is necessary to develop IEMs which can work stably at high temperature (e.g., above 100 °C) (Douglin et al., 2020). In addition, alkaline solution is usually used to clean IEMs to prevent membrane fouling, and therefore it is important to develop IEMs which is stable in alkaline environment (Alam et al., 2021).

As shown in Fig. 3, the working principle of IEMs is quite different to that of ion exchange resins (IERS) which work by exchanging ions. For example, a sodium-based cation exchange resin (CER) can be used to remove calcium ions and magnesium ions from hard water to reduce hardness, and the underlying mechanism is that sodium ions and calcium/magnesium ions are exchanged. However, this is not the case for IEMs. The name itself is quite misleading as it seems that the IEMs process is also an ion exchange process. Nevertheless, the working principle of IEMs is not an ion exchange process. The basic working principle of IEMs is permselectivity. It should also be noted that, for IEMs to work properly, the ions should have the ability to move freely, and therefore directional movement of ions can be achieved, that is why in common fuel cell applications, humidification is necessary to promote ion movement and transport (Wilberforce et al., 2019). Ideally, CEMs only allow cations to pass through while AEMs only allow anions to pass through, which corresponds to perfect permselectivity. Perfect permselectivity is desirable. However, it is very challenging to achieve this in practice because membrane permselectivity is strongly affected by other membrane properties, such as water content (or water uptake).

**Table 2**  
Summary of some reported MMMs in recent years.

Reference	Nanofiller	Polymer matrix	Main findings
Sheng et al. (2020)	ZSM-5 zeolite	Polyvinyl alcohol (PVA)	Selective removal of monovalent cations using MMM was improved.
Li et al. (2019)	Carbon nanotubes	Sulfonated poly(ether ether ketone) (SPEEK)	The MMM showed improved vanadium redox flow battery (VRFB) cell performance.
Hosseini et al. (2017b)	Graphene oxide	Polyvinyl chloride (PVC)	Membrane selectivity and electrical resistance of the MMM were comparable to commercial heterogeneous CEMs.
Barbar et al. (2019)	Silica (SiO <sub>2</sub> )	Sulfonated polyethersulfone (SPES)	The MMM showed enhanced removal of Pb <sup>2+</sup> and Cd <sup>2+</sup> ions from aqueous solutions.
Imran et al. (2020)	Titanium dioxide (TiO <sub>2</sub> )	Sulfonated polybenzimidazole (SPBI)	The MMM was promising for high temperature PEMFC applications.
Imaan et al. (2021)	Zinc oxide (ZnO)	PVA	The MMM showed promising properties for direct methanol fuel cell applications.
Hosseini et al. (2016)	Clay nanoparticle	PVC	The MMM was promising for ED applications.

Generally, a higher water content will result in a higher membrane swelling, which will negatively affect membrane dimensional stability. Dimensional stability has a strong effect on membrane permselectivity. If the dimensional stability is decreased, so will the membrane permselectivity. Although decreasing surface hydrophilicity could decrease water content and therefore improve membrane dimensional stability, it could also increase membrane resistance. A higher membrane resistance corresponds to a lower conductivity, which is not desirable as it will decrease energy efficiency and increase cost (Lei et al., 2021; López-Cázares et al., 2018; Yang et al., 2020). However, in order to achieve high ion conductivity, membrane permselectivity will usually be compromised. This tradeoff relationship between membrane permselectivity and conductivity is a critical challenge and more research is needed to address this issue, such as by exploring novel and emerging membrane materials with desired properties (Fan et al., 2020b).

**Table 3**  
Electrochemical properties of some reported IEMs and commercial IEMs.

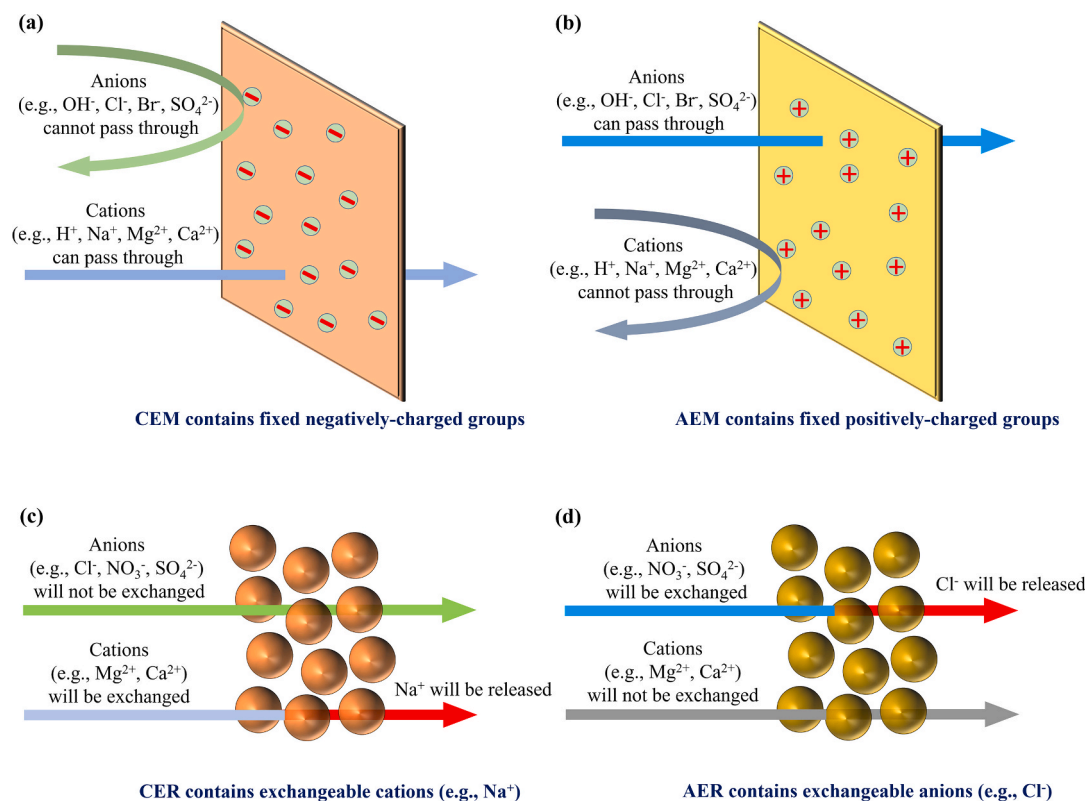
Membrane	Type	Reference/Manufacturer	Electrical resistance ( $\Omega \text{ cm}^2$ )	Permselectivity (%)	IEC (meq/g)
S5	CEM	Hosseini et al. (2017a)	4–5	>99	1.59
sCNT 10 w/w%	CEM	Fan et al. (2020a)	1.31 ± 0.03	86.2	1.66
S/P K90	CEM	Zhao et al. (2020)	1.6–3.3	94	0.60–0.61
PVA-20%	CEM	Dong et al. (2021)	2.55	90.4	1.8
PPO-PVA 1	CEM	Hong et al. (2019)	1.54	87.3	1.91
PDDA-PVA 3	AEM	Hong et al. (2019)	0.71	59.5	1.5
PPO-DMODA-3	AEM	Wei et al. (2020)	2.78 ± 0.34	94.6 ± 0.6	1.71 ± 0.07
Neosepta CMX	CEM	Tokuyama Co., Japan	2.91	99	1.62
Neosepta AMX	AEM	Tokuyama Co., Japan	2.35	91	1.25
FKD	CEM	FumaTech GmbH, Germany	<3	>94	1.2–1.4
FKS (no reinforcement)	CEM	FumaTech GmbH, Germany	0.9–1.9	98–99	1.3–1.4
FAS (no reinforcement)	AEM	FumaTech GmbH, Germany	0.4–0.8	94–97	1.6–1.8
Ralex® CMH-PES	CEM	MEGA, Czech Republic	<18	>90	2.2–2.3
Ralex® AMH-PES	AEM	MEGA, Czech Republic	<7.5	>90	1.8–2.0
AMI-7001S	AEM	Membranes International Inc., USA	<40	90	1.3 ± 0.1
CMI-7000S	CEM	Membranes International Inc., USA	<30	94	1.6 ± 0.1

### 3. Synthesis of IEMs

The most common technique for making membranes is solution casting followed by phase inversion (Li et al., 2016; Sinha and Purkait, 2015; Yun et al., 2006), which can be further divided into three main types, including thermally induced phase separation (TIPS) (Wang et al., 2017), vapor-induced phase separation (VIPS) (Zhao et al., 2018), and non-solvent induced phase separation (NIPS) (Liu et al., 2019; Susanto et al., 2019; Tan and Rodrigue, 2019). For the TIPS process, solution is prepared by dissolving polymer in a solvent at an elevated temperature, and then the solution is cooled down to induce phase separation and polymer solidification, and then the solvent is removed via solvent exchange to form microporous membrane (Cheng et al., 2014; Wang et al., 2017). TIPS is usually used to prepare membranes when a suitable solvent is not available at room temperature (Wang et al., 2017). For the VIPS process, the wet film is exposed to a gaseous non-solvent atmosphere for a period of time which can reduce mass transfer rate between the solvent and non-solvent, and then immersed in a non-solvent bath to get the membranes (Zhao et al., 2018; Zhu et al., 2018). VIPS is used to tailor membranes with desired pore structure and morphologies. For the NIPS process, a polymer solution is prepared and then is cast into a thin film, and then the wet film is immersed in a non-solvent bath to induce phase separation (Wang et al., 2019). Membrane pore size can be effectively controlled via NIPS but it is not easy to precisely control the phase separation process (Tan and Rodrigue, 2019). For synthesis of IEMs, usually NIPS is adopted. In addition, after solution casting, IEMs can also be produced by solvent evaporation to provide a dense and nano-porous structure, and usually volatile solvents are used and/or heating is applied to accelerate the evaporation process (Avcı et al., 2020b). However, one critical issue for solution casting technique is that organic solvent is usually unavoidable (Table 5), which could pose threat to the environment as well as human health as many of the organic solvents are carcinogenic. In addition, post-disposal of the organic solvent is necessary, which also inevitably increases production cost. In order to solve this issue, several strategies are developed and/or

**Table 4**  
Types of IEMs based on operating conditions.

Operating condition/environment	Type of IEM	Example
High temperature	High temperature stable IEM	Douglin et al. (2020)
Alkaline environment	Alkaline stable IEM	Alam et al. (2021)
Acidic environment	Acid stable IEM	Das and Shahi (2020)
High-concentration industrial wastewater	Antifouling IEM	Li et al. (2021)
Chlorine exposure	Chlorine resistant IEM	Mei et al. (2018)



**Fig. 3.** Schematic illustration of the working principle of IEMs and IERs. (a) Working principle of cation exchange membrane (CEM). (b) Working principle of anion exchange membrane (AEM). (c) Working principle of cation exchange resin (CER). (d) Working principle of anion exchange resin (AER).

being developed to avoid the use of organic solvents, such as to use water to replace organic solvents (L. Wang et al., 2017), in-situ polymerization of monomers containing functional groups (Hao et al., 2013), and hot-pressing (Krishna et al., 2021). When choosing the hot-pressing method, it is important to control the temperature probably due to the presence of functional ionic groups in the polymer which is thermally unstable at high temperatures.

As demonstrated in Fig. 4 and revealed by Table 5, sulfonation is usually involved in the synthesis of CEMs to introduce functional sulfonic groups into the polymer matrix, where concentrated sulfuric acid or chlorosulfonic acid are usually used as the sulfonating agent. However, both acids are strongly corrosive and dangerous. Meanwhile, a large amount of acid waste is produced after sulfonation, which is hazardous to the environment and should be disposed carefully and properly. As a result, the conventional sulfonation process becomes time-consuming and cost-ineffective. There are strategies to solve this problem, e.g., to develop more safe and green sulfonating agents such as gas SO<sub>3</sub>, to skip the sulfonation step directly by using monomers containing sulfonate groups, and to explore novel functional groups to replace sulfonate groups for CEMs. Furthermore, it is worth mentioning that linear sulfonated polymers are typically hydrophilic and tend to dissolve in water. To solve this issue, the polymers are usually cross-linked, which can make the polymers insoluble in water and improve membrane dimensional stability (Fig. 4(c)). For synthesis of AEMs, there are usually two successive steps, including chloromethylation followed by quaternization to introduce quaternary ammonium groups. However, during these processes, many toxic chemicals are used. For example, chloromethyl methyl ether (CMME), which is a common reagent for chloromethylation, is carcinogenic to human beings. Although CMME is replaced by other chemicals in some studies, such as paraformaldehyde (Baghodrat et al., 2020) and chlorotrimethylsilane (Duan et al., 2018), these chemicals are only less dangerous compared to CMME but still harmful to human health. A better solution to this issue is to bypass the

chloromethylation process through direct quaternization of nitrogen-containing polymers (Qaisrani et al., 2018). Although there are various synthesis methods, green synthesis of IEMs has a more promising future due to the growing concern of environmental pollution and sustainable development. However, though many methods are reported or being developed aiming to synthesize IEMs greenly, most of these methods can only be classified as partially green. For example, novel IEMs are synthesized using chitosan, which is regarded as environmentally friendly and sustainable as it is derived from renewable resources (Ryu et al., 2019). Compared to IEMs synthesized using traditional polymers derived from fossil fuels, the use of chitosan is a big advancement. However, organic solvents are still used during the preparation process. More efforts are needed in future research to synthesize IEMs in a completely green and sustainable way, e.g., the raw materials are derived from renewable sources, the synthesis process involves no harmful chemicals and organic solvents, etc.

#### 4. Applications of IEMs

IEMs are extensively applied in various applications, which can be generalized into two big categories, where the water-based category mainly includes electro dialysis, diffusion dialysis and membrane capacitive deionization, while the energy-based category mainly includes reverse electro dialysis, fuel cells, redox flow battery and electrolysis. These applications are discussed in detail below.

##### 4.1. Electrodialysis

Electrodialysis (ED), as a separation process driven by electricity, is one of the most important applications of IEMs (Al-Amshawee et al., 2020; Campione et al., 2018; Ran et al., 2017). Schematic illustration of the ED process is shown in Fig. 5(a). An ED cell usually has two half parts where each part has an electrode, either anode or cathode. Before the

**Table 5**  
Recent studies on synthesis of CEMs.

Reference	Main solvents	Main synthesis procedures
Kang et al. (2015)	DMAc, DMSO	(1) Polyvinylidene fluoride (PVDF) is dehydrofluorinated; (2) polymerization of sodium styrenesulfonate (NaSS) takes place on PVDF in dimethyl sulfoxide (DMSO); (3) PVDF-grafted-polystyrene sulfonate (PSS) is dissolved in dimethylacetamide (DMAc), and the solution is casted on a film; (4) the membrane is annealed at various temperatures.
Reig et al. (2015)	NMP	(1) PVDF is sulfonated using HSO <sub>3</sub> Cl; (2) sulfonated PVDF (S-PVDF) and PVDF are dissolved in N-methyl-2-pyrrolidone (NMP) separately; (3) the S-PVDF/PVDF blended solution is casted on a glass plate and dried; (4) membrane surface is modified (e.g., via polymerization of aniline) to improve monovalent cation selectivity.
Martos et al. (2017)	DMAc, Toluene	(1) Copolymers of polyether ether ketone (PEEK) and polyether sulfone (PES) containing cardo (lactone) group are synthesized in DMAc and toluene; (2) copolymers are sulfonated using HSO <sub>3</sub> Cl; (3) sulfonated and non-sulfonated copolymers are dissolved in DMAc and casted on a glass plate and dried.
Kim et al. (2017a)	DMSO	(1) Polyketone solution is casted on a glass plate and dried; (2) polyketone membrane is immersed into NaSS-DMSO solution and exposed to irradiation to initiate the grafting reaction.
Rikame et al. (2017)	Toluene, CHCl <sub>3</sub> , H <sub>2</sub> O	(1) Fullerene is phosphorylated; (2) polyvinyl alcohol (PVA) is sulfonated using HSO <sub>3</sub> Cl and then the S-PVA solution is casted on a glass plate and dried; (3) phosphorylated fullerene powder is added to S-PVA solution, then the mixture is casted on a glass plate and dried.
Namdari et al. (2017)	THF	(1) Polyvinyl chloride (PVC), high impact polystyrene, and acrylonitrile butadiene styrene with different ratios are dissolved in tetrahydrofuran (THF); (2) cation exchange resin (CER) and nanoparticles are added into the polymer solution, and then the mixture is casted on a glass plate, and then dried and immersed in water to get the membrane.
Petreanu et al. (2017)	CHCl <sub>3</sub> , DMF	(1) Polydimethyl phenylene oxide (PPO) in dissolved in CHCl <sub>3</sub> , and is sulfonated using HSO <sub>3</sub> Cl; (2) S-PPO is dissolved in dimethylformamide (DMF), and then casted and dried; (3) tetraethyl orthosilicate is added to S-PPO solution, and the mixture is casted on a glass plate and dried to get the composite membrane.
Hosseini et al. (2018)	Methanol, THF	(1) Zn(II) nanoparticles are prepared; (2) PVC is dissolved in THF; (3) CER and Zn(II) nanoparticles are added to the PVC solution, and then the mixture is casted on a glass plate, and then dried and immersed in water to get the membrane.
Li et al. (2017)	NMP, DMSO, Methanol	(1) PVDF is dissolved in NMP and NaSS is dissolved in DMSO; (2) PVDF-NMP solution undergoes ozone treatment via O <sub>3</sub> /O <sub>2</sub> ; (3) the PVDF solution and NaSS solution are mixed and polymerization takes place, producing the PVDF-g-PSS copolymers; (4) PVDF-g-PSS is dissolved in NMP, and then casted onto a glass plate, and then dried and immersed in water to get the membrane.
Zhao et al. (2018)	DMAc	(1) PES is sulfonated using concentrated H <sub>2</sub> SO <sub>4</sub> ; (2) the sulfonated PES is dissolved in DMAc; (3) polyvinyl pyrrolidone is added into the solution; (4) the solution is casted on a glass plate, and then dried and immersed in water to get the membrane.

**Table 5 (continued)**

Reference	Main solvents	Main synthesis procedures
Rajput et al. (2018)	Cyclohexanone	(1) Graphene oxide is prepared and sulfonated using concentrated H <sub>2</sub> SO <sub>4</sub> ; (2) PVC is dissolved in cyclohexanone; (3) styrene and divinylbenzene are added to the PVC solution and then undergo polymerization; (4) after reaction, the product is casted on a glass plate and dried; (5) the membrane is sulfonated using HSO <sub>3</sub> Cl; (6) sulfonated graphene oxide is dissolved in cyclohexanone, and then added into the copolymer solution, and then the resulting solution is casted on a glass plate, dried and sulfonated to get the composite membrane.
Chen et al. (2019)	NMP	(1) Copolymer of sulfonated polybenzimidazole (PBI), PBI containing pyridine, and polyimide are synthesized; (2) the copolymer is dissolved in NMP, and then the solution is casted on a glass plate, dried and immersed in water to get the membrane; (3) the membrane is immersed in phosphoric acid to get the proton exchange membrane (PEM).

two parts are fastened together, a set of alternating cation exchange membranes (CEMs) and anion exchange membranes (AEMs), which are separated by spacers, are placed between the two parts. AEMs act as barriers for cations while CEMs act as barriers for anions. The anode and cathode of the ED cell are then connected to a power source. Besides, the cell also has inlets and outlets which are connected to three external compartments, including the electrode compartment, concentrating compartment, and diluting compartment. Usually, the solutions are pumped into the cell where solution in the diluting compartment will be desalted while salt concentration in the concentrating compartment will be increased (Hosseini et al., 2020). It should be emphasized that AEMs and CEMs should be carefully arranged in an ED cell to ensure that when power is supplied, cations in the concentrating compartment will not move towards CEMs and anions in the concentrating compartment will not move towards AEMs.

In addition to conventional ED, electrodeionization (EDI) is an ED-derived technology that combines ion exchange resins together. The main difference between an EDI cell and ED cell lies in that ion exchange resins are integrated into the diluting compartment in an EDI cell. EDI technology can reduce the undesired concentration polarization occurred in an ED process (Alvarado and Chen, 2014; Jin et al., 2016). Meanwhile, for EDI process, no additional chemicals are needed to regenerate ion exchange resins, which is a main advantage compared to standalone ion exchange technology. Furthermore, the ion exchange resin in the diluting compartment can reduce the resistance of the EDI cell, especially when the salt concentration in the diluting compartment becomes very low (Alvarado and Chen, 2014). EDI can produce very high purity water and is often used to polish reverse osmosis permeate (Alvarado and Chen, 2014; Li et al., 2017). However, the use of loose resin beads in EDI could result in unstable performance and channel formation. Therefore, it is important to develop methods to immobilize the loose resin beads, which can be done by a resin wafer with porous and solid matrix structure containing immobilized resin beads (Pan et al., 2018).

ED is widely used for seawater desalination (Doornbusch et al., 2019, 2020), brackish water desalination (Chen et al., 2020; Wright et al., 2018), water and wastewater treatment in various industries, such as paper industry and electroplating industry (Barros et al., 2020; Bdiri et al., 2018; Nataraj et al., 2007; Ye et al., 2020). For example, Doornbusch et al. (Doornbusch et al., 2019, 2020) investigated the practicality of ED for seawater desalination from the perspective of energy consumption. The energy consumption was close to that of

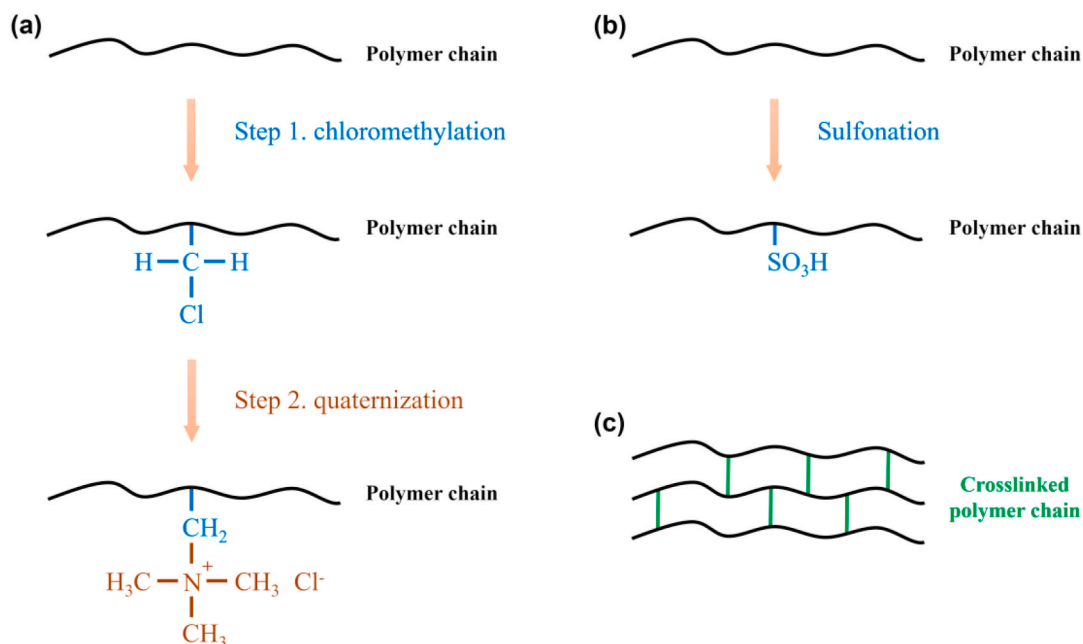


Fig. 4. Typical routes for synthesis of IEMs. (a) Typical route for synthesis of AEMs. (b) Typical route for synthesis of CEMs. (c) Crosslinked polymer chain.

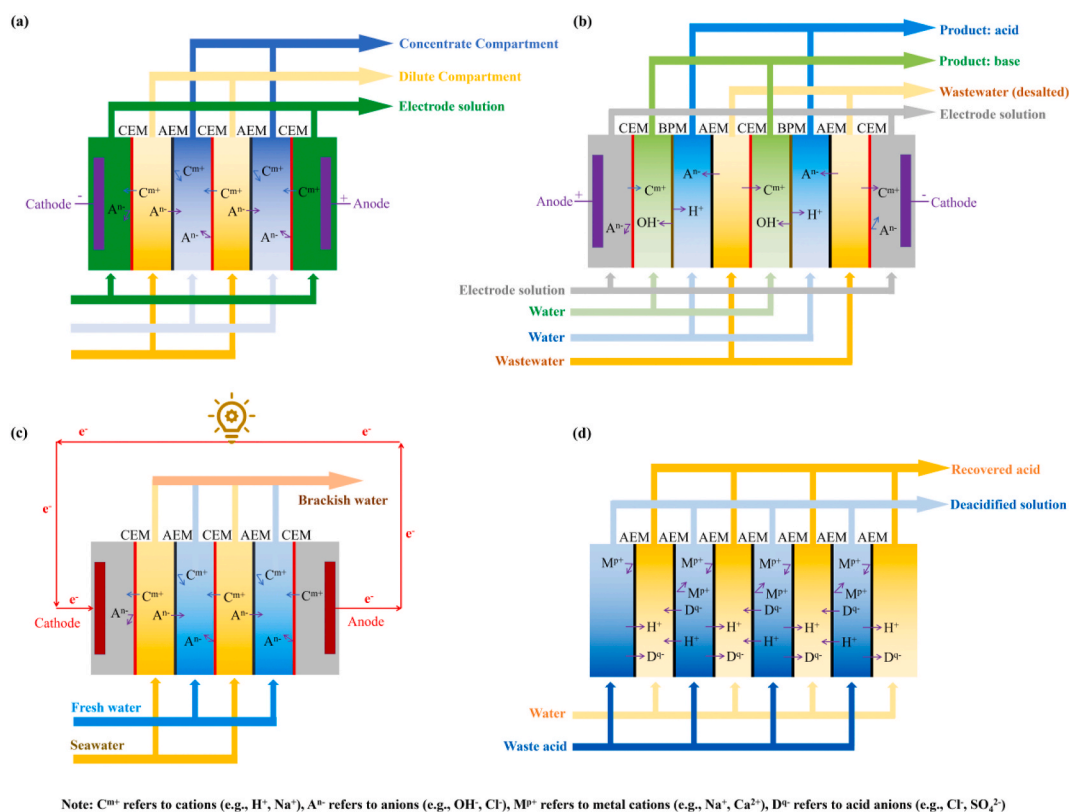


Fig. 5. Schematic illustration of a typical ED process (a), BMED process for acid and base recovery (b), RED process for electricity production (c), and diffusion dialysis process for acid recovery (d). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

state-of-the-art reverse osmosis desalination technology (Doornbusch et al., 2019). However, it is impossible to achieve full desalination in a single stage so multiple stages are necessary. Xu et al. (2018a) conducted bench and pilot-scale studies of brackish groundwater desalination using ED technology and they investigated the effects of operating conditions such as current density, linear velocity, hydraulic retention

time, and staging on desalination performance. Especially, they found that surface coating was an effective method to increase membrane selectivity between monovalent cations and divalent cations. Zhang et al. (2017) designed an ED system to treat seawater concentrate, which was produced by reverse osmosis plant, to produce coarse salt using monovalent selective IEMs under continuous operation. Alternatively,

the concentrated brine after ED treatment can also be used as raw material for chlor-alkali industry (Reig et al., 2014). Shah et al. (2019) designed a feed-forward voltage-controller which would benefit ED system designers and operators since it could be used in batch ED system to improve desalination performance by changing voltage.

ED technology is also used in food industry (Bhattacharjee et al., 2017; Kravtsov et al., 2020; Wang et al., 2020). For example, for fruit juice production, it is important to reduce the amount of organic acids that could be present in the juice, since these acids can cause side effects to human bodies and can also affect the taste (Faucher et al., 2018). ED has proved to be an effective technology to deacidify fruit juice, such as cranberry juice (Pelletier et al., 2017; Serre et al., 2016). In addition, other ED-based food industry applications include but not limited to dairy (Talebi et al., 2019), wine (Bdiri et al., 2020), and wastewater treatment (Bdiri et al., 2018). However, wastewater from food industry usually have high concentrations of organic and inorganic matters, colloidal particles, microorganisms and macromolecules (Bdiri et al., 2019). These complex components in food industry solutions make membrane fouling easier to occur during ED process, which reduces the lifetime of IEMs and increases operation cost. More generally, it is easier for fouling to occur when feedwater has complicated compositions and high concentrations. Therefore, more research is needed to develop excellent antifouling IEMs which can work stably and consistently in harsh environment (Al-Amshawee et al., 2020).

Bipolar membrane electrodialysis (BMED) is a special type of ED which involves using bipolar membranes. A typical BMED process for acid and base recovery is illustrated in Fig. 5(b). One of the most obvious characteristics of BMED technology is that hydrogen ions and hydroxide ions can be efficiently produced through water dissociation at the interface of the cation exchange layer (CEL) and anion exchange layer (AEL) of the membrane (Pärnamäe et al., 2021b; van Linden et al., 2020). Therefore, BMED is widely used in acid and base production from salt solutions (Achoh et al., 2019; Ashrafi et al., 2017; Shi et al., 2018; Sun et al., 2017). In addition, BMED is also a favourable technology in food industry as no chemical additives are produced during food processing via BMED (Li et al., 2020). A lot of studies are carried out on BMED. It was found that BMED was effective in simultaneously separation and recovery of lithium (recovered as LiOH) and boron (recovered as  $H_3BO_3$ ) from aqueous solution (Bunani et al., 2017; İpekçi et al., 2018). Zhang et al. (2019) adopted ion exchange technology to remove calcium ions and magnesium ions from desulfurization wastewater, and then the desulfurization wastewater was further treated via BMED process, during which high-purity acid and base were produced. Szczygielida et al. (Szczygielida and Prochaska, 2017) developed a BMED process to produce alpha-ketoglutaric acid and they found that high concentration of alpha-ketoglutaric acid, high current efficiency, and low energy consumption could be achieved simultaneously under optimized conditions. Lv et al. (2018) recovered hydrochloric acid and ammonium hydroxide from ammonium chloride wastewater using BMED technology and the process cost could be decreased by optimizing operation conditions such as current density and initial concentration of ammonium chloride. BMED is also promising in recovering citric acid from fermented liquid due to its low energy consumption and high recovery rate of citric acid under optimized conditions (Sun et al., 2017), and could also be employed to effectively recover ammonium ions, phosphate ions, and volatile fatty acids from pig manure hydrolysate (Shi et al., 2018). However, as a bipolar membrane consists of two separate layers which contain oppositely charged groups, it is challenging to maintain the stability of the layers, and especially, the AEL layer of the membrane is not stable in alkaline conditions (Giesbrecht and Freund, 2020). As a result, it is challenging to maintain consistent operating performance. Therefore, future research is needed to develop more durable and stable membranes.

In addition to IEM-based water treatment technologies (e.g., ED), there are also many other technologies for water treatment, including but not limited to sand filtration (Xu et al., 2019),

coagulation-flocculation-sedimentation (Zhang et al., 2019), activated carbon adsorption (Cermakova et al., 2017), ion exchange resin (Amini et al., 2015), thermally-driven membrane distillation technology (Han et al., 2017), low-pressure-driven microfiltration and ultrafiltration membrane technologies (Szymański et al., 2019; Waeger et al., 2010), high-pressure-driven nanofiltration and reverse osmosis membrane technologies (Adam et al., 2018; Liu et al., 2011), osmotically-driven forward osmosis membrane technology (Chen et al., 2019), disinfection technologies such as UV irradiation and chlorination (Al-Gabr et al., 2013; Zhang et al., 2019), advanced oxidation technologies such as electrochemical advanced oxidation process (Brillas, 2020), and biological treatment technologies (Berillo et al., 2019; Marsidi et al., 2018). Furthermore, in recent years, there are also some emerging and promising chemicals, materials and technologies for water and wastewater treatment, such as metal organic frameworks (MOFs) based mixed matrix membranes (Jun et al., 2020a; Kim et al., 2020), MXene (Jun et al., 2020b), activated persulfate for bisphenol A removal (Jhones dos Santos et al., 2021), etc. Especially, MXene, as a very recently developed new family of two-dimensional materials, has excellent properties such as oxidation resistance and stability, and demonstrates high adsorption performance for selected pharmaceuticals and dyes (Jeon et al., 2020; Jun et al., 2020c; Kim et al., 2021). MXene can also be used to prepare filtration membranes which show enhanced water flux and antifouling performance (Al-Hamadani et al., 2020).

The above technologies can be divided into membrane technologies and non-membrane technologies. Generally, membrane technologies are more advanced water treatment technologies and can produce water with high quality and purity (Zheng et al., 2015). The main advantages and disadvantages of different membrane technologies are summarized in Fig. 6 (Al-Amshawee et al., 2020; Xu et al., 2017; Zou et al., 2019). It should be clarified that the advantages and disadvantages are not absolute. For example, compared to reverse osmosis (RO), ED has the advantage of low membrane fouling, however, it does not mean that fouling will not easily occur in an ED system. In fact, as discussed earlier, fouling can occur under certain conditions. As the cost of membrane technologies has decreased significantly in recent years, they are being widely used in water treatment. In fact, membrane technologies are employed in most of the desalination plants today (Al-Amshawee et al., 2020). Compared to other water treatment technologies, ED is especially effective and efficient in removing ions from water. Therefore, ED can be used as a pretreatment technology for RO process to reduce RO membrane scaling. Also, ED can be used to treat RO concentrate. Generally, the compositions of real industrial wastewater can be very complex and diverse. However, ED cannot remove other water pollutants such as suspended particles, colloids, bacteria, and large organic molecules. Therefore, in practice, in order to get high purity water, other water treatment technologies are needed to remove these pollutants. Integration of ED with other water treatment technologies have been increasingly reported in various operation ways. For instance, Cui et al. (2017) investigated an integrated pilot-scale system of electrochemical oxidation, upflow biological aerated filter, and ED for advanced treatment of triazole fungicides discharged water. In this system, electrochemical oxidation was applied as pretreatment process to enhance the wastewater biodegradability for the following treatment of upflow biological aerated filter, and then ED process was applied for salt removal. Differently, in a groundwater recharge program of Belgium, ED was integrated in the first place to reduce the salinity of RO concentrate, followed by ozonation and biological treatment to remove organic compounds from ED effluent (Zhang et al., 2011). Furthermore, integration of ED before ozonation can remove bromide to relatively low concentration, and thus bromate formation will not be a problem during subsequential ozonation treatment (Van der Hoek et al., 2000).

#### 4.2. Diffusion dialysis

As shown in Fig. 5(d), in a diffusion dialysis (DD) cell, there are



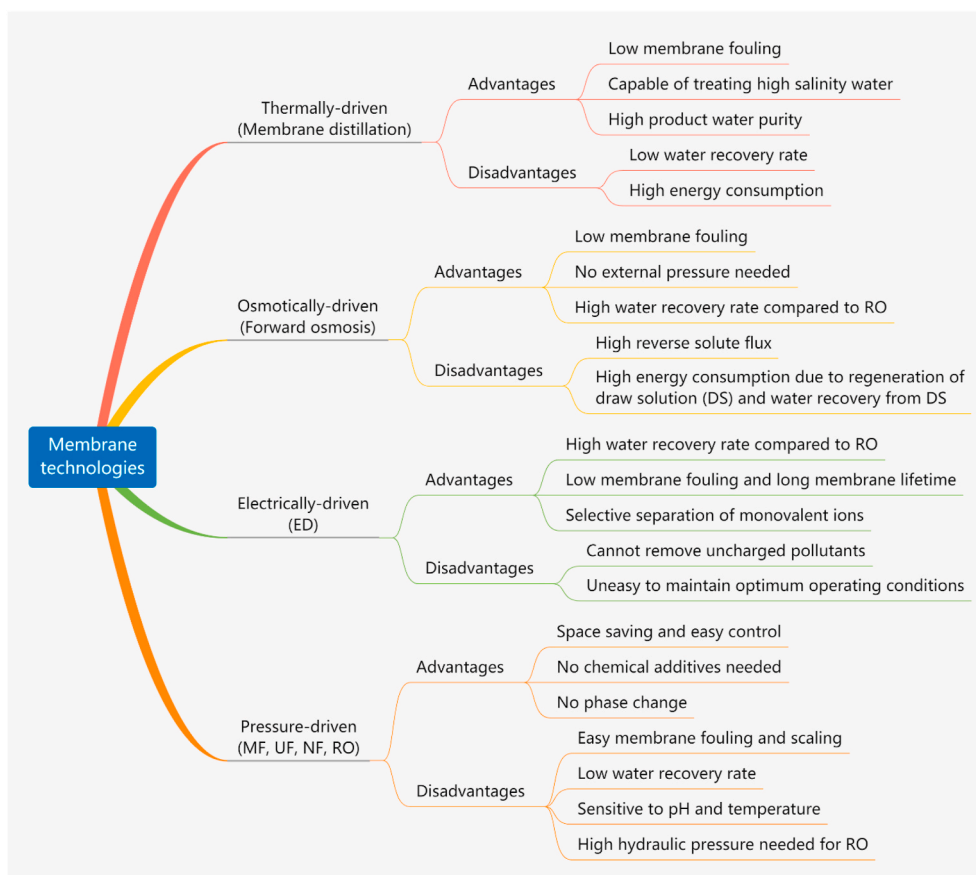


Fig. 6. Comparison of common membrane technologies for water treatment. MF refers to microfiltration; UF refers to ultrafiltration; NF refers to nanofiltration; RO refers to reverse osmosis; ED refers to electro dialysis.

commonly two different parts with different concentrations which are separated by IEMs. Driven by concentration difference, ions can diffuse through the membrane from the high concentration solution part to the low concentration solution part. Small counter-ions will also pass through the membrane to balance the charges. DD is a spontaneous process which virtually does not need external energy input (Bendová and Palatý, 2018). Therefore, it is a process with low cost and low energy consumption (Wang et al., 2018). DD is regarded as a promising method for acid recovery (Emmanuel et al., 2016; Lin et al., 2017). However, the DD is very slow and inefficient process because the driving force is very weak (Luo et al., 2011). Although large membrane area can be applied to speed up the process, it will increase the capital cost, rendering DD less favourable. Therefore, future research is needed to improve the acid recovery capacity and efficiency, which can be done by developing thin and porous membranes to reduce ion transport resistance (Lin et al., 2021).

#### 4.3. Membrane capacitive deionization

For membrane capacitive deionization (MCDI), CEMs and AEMs are used simultaneously (Fig. 8(d)). The cations from the salted water (e.g., brackish water) arrive at the CEM side, pass through the CEM and are absorbed by the porous electrode which is usually made of carbon materials such as carbon nanotubes and carbon aerogels (Li and Zou, 2011). In contrast, the anions from the salted water arrive at the AEM side, pass through the AEM and are absorbed by the porous electrode. MCDI technology combines the advantages of capacitive deionization (CDI) technology and IEM technology. Compared to CDI, MCDI can remove ions from water more efficiently and is regarded as a promising technology for brackish water desalination. Salt removal efficiency during

MCDI process is strongly affected by membrane resistance/conductivity and a lot of studies are focused on developing IEMs with low resistance (Palakkal et al., 2018). Zhang et al. (2015) prepared a series of novel AEMs and they found that by incorporating reduced graphene oxide/polyaniline into the membranes, the electrical conductivity of the membranes was improved greatly, and the salt removal efficiency and adsorption capacity during MCDI process increased greatly. Kim et al. (2019) developed novel pore-filled CEMs which demonstrated high selectivity coefficients for multivalent cations, and during MCDI process using these membranes, high removal efficiency for multivalent cations and high energy recovery efficiency were achieved. Qiu et al. (2017) prepared very thin CEMs that had very low electrical resistance, and the current efficiency during MCDI process using these membranes was comparable to that using commercial membranes. Kim et al. (2017b) developed CEMs via radiation grafting technique and during MCDI process using these membranes, the salt removal rate was about three times as fast as that of CDI process. However, as MCDI is still at its early stage, more research is needed to gain a better understanding of the process and optimize the parameters. For example, unlike electro dialysis where IEMs are used to separate the compartments, IEMs for MCDI application just function as a conformal layer for ion selectivity, therefore membrane thickness for MCDI can be optimized (Ali et al., 2020). Also, more efforts are needed to improve the energy efficiency of MCDI by developing IEMs with high ionic conductivity and integrating them with the electrodes.

#### 4.4. Reverse electro dialysis

Though reverse electro dialysis (RED) contains the word “electro dialysis”, it is quite different from ED. As discussed above, ED is mainly

used in water treatment to remove ions from water, and electrical energy is consumed in an ED process. However, RED is used to produce energy (Cho et al., 2017a; Farrell et al., 2017). In terms of driving force, ED is driven by external power source while RED is driven by concentration difference or salinity difference. Therefore, RED usually involves mixing two aqueous solutions with different salinities (Jande and Kim, 2014; Zlotorowicz et al., 2017). The energy arisen from this salinity difference is known as salinity gradient power (Mei et al., 2018). To be more specific, due to salinity difference, seawater and river water/freshwater possess different electrochemical potentials. As ion transport happens between the two aqueous solutions via IEMs, the electrochemical potentials are reduced (Cho et al., 2017b). In the meantime, power is generated. Schematic illustration of RED process for power generation is shown in Fig. 5(c). RED is a promising renewable energy technology for large-scale power generation (Hong et al., 2015). High power densities could be obtained from RED by changing cell design with particular focus on membrane resistance and the cell length (Vermaas et al., 2012). It should be noted that reverse electrodialysis (RED) and electrodialysis reversal (EDR) are two different terms. EDR refers to reversing the polarity of the electrodes at regular time intervals and EDR is developed to reducing membrane fouling (Zhao et al., 2019). A lot of studies on RED have been carried out. For example, as seawater and river water contain multivalent ions which would limit RED performance, researchers developed a method to improve membrane selectivity towards monovalent ions and multivalent ions by coating a layer on commercial membranes (Güler et al., 2014). Fontananova et al. (2017) investigated the effect of solution concentration and composition on ionic resistance and permselectivity of membranes which would affect power density obtained from RED. Fan et al. (Fan and Yip, 2019) developed a membrane transport model to elucidate the tradeoff relationship between conductivity and permselectivity in ED and RED. They concluded that using hypersaline streams of saltworks brine and seawater concentrate for power generation via RED would not be feasible due to the significantly diminished permselectivity which could reduce energy extraction efficiency. Currently, RED is not a cost-effective technology and there are also some technical issues which hinders the large-scale application of RED. Therefore, more efforts are needed to lower operation cost and improve power output, where one strategy is to innovatively redesign the RED stack components and system (Mei and Tang, 2018). Furthermore, more efforts are needed to develop RED-customized IEMs with high permselectivity and low electrical resistance, which can accelerate the commercialization of RED (Hong et al., 2019).

#### 4.5. Fuel cells

Fuel cell, an electrochemical device which can convert chemical energy of fuels into electrical energy without combustion, is another common application of IEMs (Jiang and Ladewig, 2017; Kamecev et al., 2017; Peighambaroust et al., 2010). Common fuels used in fuel cells include but are not limited to hydrogen, methanol, ethanol, and natural gas (Alias et al., 2020; Greene et al., 2020; Kakaei and Rahnavardi, 2021; Skabelund et al., 2020). Among the different types of fuel cells, proton exchange membrane fuel cell (PEMFC) is the most popular one (Firouz Tadavani et al., 2018; Ozden et al., 2018; Poulsen et al., 2017; Wilberforce et al., 2019). Schematic illustration of the PEMFC process is shown in Fig. 7(a). Currently, PEMFC is an expensive technology due to the high production cost of high purity hydrogen. Impurities in hydrogen, such as carbon monoxide, can deactivate the catalytic electrode materials such as platinum in PEMFC (Rosli et al., 2017). Therefore, the impurities must be removed from the hydrogen feed. In addition, although the commonly used Nafion and other commercial PEMs have an excellent proton conductivity, they cost a lot and are not stable at temperatures over 80 °C (Shabani et al., 2020). Operating fuel cells at high temperature can improve the overall performance by solving some critical problems of conventional fuel cells, such as the water flooding issue and the catalyst poisoning problem (Authayanun et al., 2015; Shabani et al., 2019). Therefore, more research is needed to develop low-cost membrane materials with excellent stability (e.g., thermal stability, chemical stability and mechanical stability) and high conductivity, and especially, to develop membranes that can function well at high temperatures (Iulianelli and Basile, 2012; Rosli et al., 2017). Currently, some attempts have been made to modify membrane materials with nanoparticles, activated carbon, clay, and bio-based carbon materials (Shabani et al., 2020). For example, Neethu et al. (2019) prepared a low cost and high-performance PEM with a price of only 45 dollars/m<sup>2</sup> by mixing activated carbon made from coconut shell with natural clay.

Anion exchange membrane fuel cell (AEMFC) is another type of fuel cell which is being actively studied in recent years. As shown in Fig. 7, water is formed at the anode for AEMFC, while for PEMFC, water is formed at the cathode. AEMFC has some attractive advantages over PEMFC, such as more flexible catalyst choices and high oxygen reduction reaction rate (Hickner, 2017; Iravaninia et al., 2017; Le Mong and Kim, 2018). However, one of the most challenging issues which hinders the commercialization of AEMFC as a competitive energy conversion system is the durability, and one possible solution from the membrane perspective is to develop novel AEMs using aryl ether-free polymer

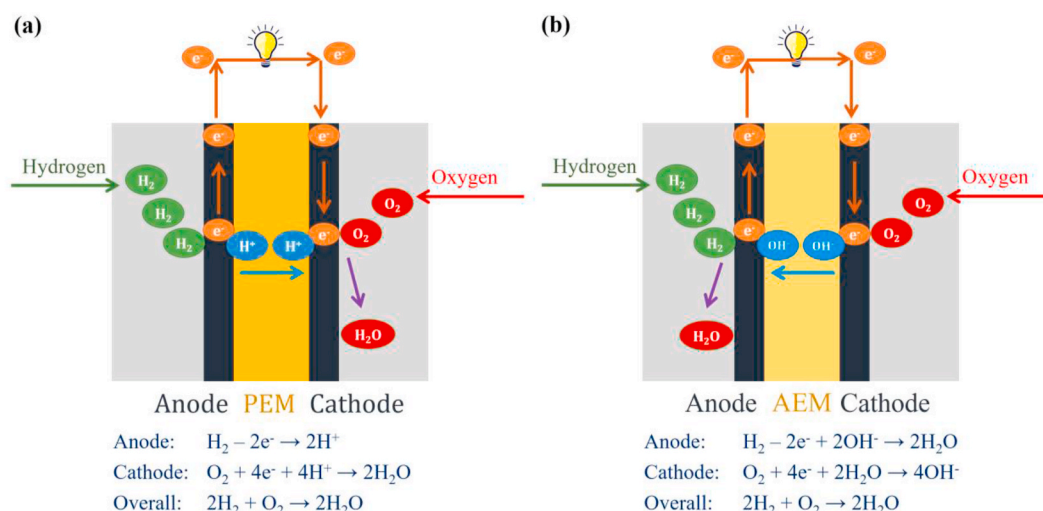


Fig. 7. Schematic illustration of a typical PEMFC (a), and AEMFC (b).

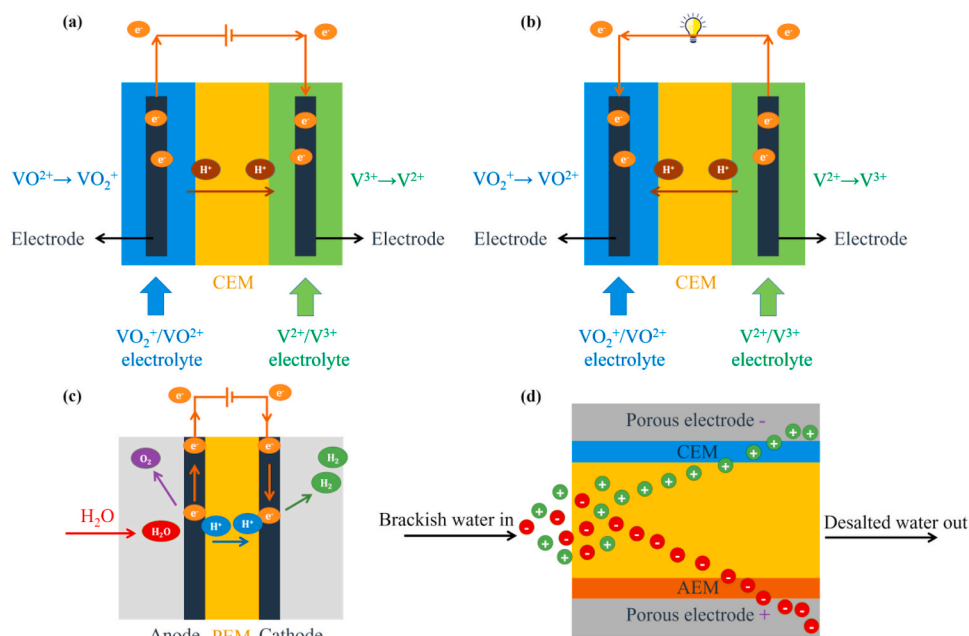
matrix with desired cationic groups, high conductivity and thermostability (Mustain et al., 2020; Ramaswamy and Mukerjee, 2019).

Another type of fuel cell involving IEMs is called microbial fuel cells (MFCs), which is an emerging technology for energy production (Leong et al., 2013; Santoro et al., 2017). Compared to conventional fuel cells, MFCs have two obvious advantages (Arun et al., 2020; Asensio et al., 2018; Munoz-Cupa et al., 2021). Firstly, it is more cost-effective because microorganism, instead of expensive catalysts (e.g., platinum), is used as the catalyst while wastewater is used as the fuel to replace conventional fuels in fuel cells. Secondly, MFCs can be applied to wastewater treatment while generating electricity at the same time. Separator material is an important factor affecting the electrical conductivity of MFCs. Inserting the separator material between the two electrodes can not only increase the concentration of the substances required for the oxidation-reduction reaction, but also improve the power generation efficiency and transfer rates (Sun et al., 2016). Although there are various separator materials (e.g., IEMs, fiber membranes, and ultrafiltration membranes), PEM with better performance is most frequently used. PEM only transmits  $H^+$  ions required for cathodic reduction reaction in one direction, so the oxygen reduction reaction rate is accelerated and the electrical performance of MFCs is improved. Another vital role of the PEM is to prevent oxygen transport from the aerated cathode to the anaerobic anode chamber (Shabani et al., 2020). However, in addition to the high cost of PEMs, the efficiency of PEMs in channeling protons could be hindered because of biofouling, oxygen crossover and substrate loss, which reduces the service life and the conductivity of the membrane (Olayiwola Sirajudeen et al., 2021). These issues must be solved in the future before MFCs can be widely commercialized.

#### 4.6. Redox flow battery

Basically, redox flow battery (RFB) works by converting chemical energy into electricity. In a RFB system, two electrolyte solutions are pumped into the regions which contain the electrodes and are separated by an IEM. Redox reactions take place in these regions. Vanadium redox flow battery (VRFB) is the most common type of RFB and the vanadium ions involved in the system include  $VO_2^+$ ,  $VO^{2+}$ ,  $V^{3+}$ , and  $V^{2+}$  (Hwang

et al., 2018; Kim et al., 2018). The charging and discharging processes of VRFB illustrated in Fig. 8(a) and (b). During the discharging process, at the cathode,  $VO_2^+$  ions gain electrons and are reduced to  $VO^{2+}$  ions. At the anode,  $V^{2+}$  ions lose electrons and are oxidized to  $V^{3+}$  ions. VRFB technology is promising for large-scale energy storage due to its low cost, long cycle life, high efficiency, easy operation, good safety, and high design flexibility compared to other technologies (Liao et al., 2017; Liu et al., 2017; Sharma et al., 2018). However, there are some critical challenges, such as the crossover effects, pumping losses, shunt current loss, limited energy density, degradation of the system due to the harsh environment, etc. More efforts are needed to address these issues (Lourenssen et al., 2019; Xing et al., 2011; Ye et al., 2015). From the membrane perspective, IEMs play a key role in VRFB applications and the system performance can be improved by developing more effective membranes. Therefore, a lot of studies are focused on IEMs for VRFB. For example, in order to have high voltage efficiency, IEMs used in VRFB should have low membrane resistance (Hwang et al., 2018). Kim et al. (2018) developed a series of IEMs for VRFB applications and one of the IEMs demonstrated better cell capacity and efficiency, as well as lower capacity decay compared to Nafion membranes. Liu et al. (2017) developed novel acid-base IEMs incorporating modified graphene oxides which showed great potential for VRFB applications. Especially, amphoteric IEMs are being widely studied in VRFB applications due to their advantages compared to conventional membranes, such as having low vanadium ion permeability while maintaining high conductivity (Hu et al., 2012). Sharma et al. (2018) prepared amphoteric IEMs based on styrene sulfonate and vinyl benzyl chloride and the membranes demonstrated lower vanadium ion permeability than Nafion membranes. Liao et al. (2017) developed amphoteric IEMs which demonstrated higher battery efficiencies and lower capacity loss compared to Nafion membranes. Wang et al. (2014) synthesized novel amphoteric IEMs with significantly low permeability of vanadium ions in VRFB due to the presence of quaternary ammonium groups in the membranes. However, IEM is still the major factor hindering the commercialization of VRFB. Currently, perfluorinated membranes are commonly used in VRFB and the price for these membranes is very high. An important future research direction is to develop low cost partially fluorinated and non-fluorinated IEMs with high ionic conductivity and low vanadium



**Fig. 8.** Schematic illustration of (a) the charging process of a vanadium redox flow battery, (b) the discharging process of a vanadium redox flow battery, (c) PEM-based electrolysis for hydrogen production, and (d) membrane capacitive deionization.

ion permeability to achieve high coulombic efficiency (Zhou et al., 2021).

#### 4.7. Electrolysis

IEM-based electrolysis is widely studied for hydrogen production. Generally, there are three main types of water electrolysis for hydrogen production, including alkaline electrolysis, proton exchange membrane (PEM) electrolysis (Fig. 8(c)), and solid oxide electrolysis (Amikam et al., 2018; Ju et al., 2018; Schiller et al., 2019). Compared to alkaline electrolysis, PEM-based electrolysis can reduce gas crossover significantly as the membrane can act to separate the anode and cathode, therefore hydrogen with high purity can be produced (Park et al., 2021). PEM-based electrolysis for hydrogen production is regarded as a clean technology because no pollutants are produced. However, currently it is not an economically competitive technology due to its high operational cost. Furthermore, compared to conventional alkaline electrolysis, PEM-based electrolysis typically uses more expensive catalytic electrode materials (e.g., platinum) (Gomez Vidales et al., 2018). Therefore, a lot of studies are focused on developing cost-effective alternatives to platinum. For example, Rozenfeld et al. (2018) synthesized an exfoliated molybdenum disulfide catalyst which contributed to the production of highly purified hydrogen. Kim et al. (Kim and Logan, 2019) prepared a blended material as cathode using Nickel powder and activated carbon to reduce cost for hydrogen production. However, the acidic operation conditions for PEM-based electrolysis limits the choice of catalysts. Recently, AEM-based electrolysis is developed for hydrogen production which can operate in high pH conditions, and therefore less expensive catalysts are available. For AEM-based electrolysis, more efforts are needed to develop membranes with high conductivities, good mechanical strength, and especially, high stability in alkaline conditions (Fortin et al., 2020).

IEM-based electrolysis is also used in chlor-alkali industry to produce chlorine, hydrogen and sodium hydroxide simultaneously. Generally, CEMs such as Nafion membranes are used to separate the two electrodes and thus prevent chlorine from reacting with hydroxide ions (Chikhi et al., 2002; Kiros et al., 2006). The chlor-alkali process is very energy-consuming and costly, so quite a few studies have been reported to address this issue. For example, Kiros et al. (2006) found that over 30% energy could be saved during electrolysis using highly active electrocatalysts as cathode materials compared to conventional electrolysis using cathode which involves high voltage-consuming hydrogen evolution reaction. Furuya et al. (Furuya and Aikawa, 2000) compared the performance of oxygen cathodes with silver catalyst and platinum catalyst in chlor-alkali electrolysis process. They found that although platinum demonstrated better catalytic activity compared to silver, it had a much shorter lifetime. Therefore, operation cost could be reduced by using oxygen cathodes loaded with silver catalyst rather than those with platinum catalyst. Otashu et al. (Otashu and Baldea, 2019) developed a dynamic model to optimize operations of chlor-alkali membrane electrolysis plant so as to reduce energy cost. This model could be used to study demand response strategies and it was possible to modulate the cell power demand rapidly without affecting cell concentration and temperature. Moussallem et al. (2012) developed a novel method to prepare high performance silver-based gas diffusion electrodes for chlor-alkali electrolysis with oxygen depolarized cathodes. Electrode thickness could be easily controlled by this method and also this method could be used for large scale production of electrodes. Jalali et al. (2009) investigated the effects of different operating conditions on cell voltage and current efficiency of chlor-alkali membrane cell. They found that current density and cell temperature were two deciding factors for cell voltage while brine concentration was the deciding factor for current efficiency. On the other hand, cost can also be reduced by extending the lifetime of IEMs and correspondingly, more research is needed to improve membrane durability in alkaline environment and resistance to chemical attack.

Electrolysis can also combine with other processes to achieve specific

requirements. Electro-electrodialysis (EED), which combines electrodialysis and electrolysis together, is a typical example. In a three-chamber EED cell, the cathode and anode are separated by a CEM and an AEM (Jaroszek and Dydo, 2016). Salt solution is pumped into the middle chamber, and the cations pass through the CEM and reach at the cathode chamber while the anions pass through the AEM and reach at the anode chamber. Meanwhile, water splitting occurs at the two electrodes. Similar to BMED, EED can also be used to produce, purify, concentrate or regenerate acids and bases. For example, Miao et al. (2018) developed a three-chamber EED cell to produce tetrabutyl ammonium hydroxide with high purity. The EED cell can also have only two chambers. In a two-chamber EED cell, the cathode chamber and anode chamber are separated by either an AEM or a CEM. Das et al. (2019a) developed a CEM-based two chamber EED cell to increase hydrogen iodide concentration in the mixture of hydrogen iodide and iodine solution. Wu et al. (2019) developed an AEM-based two-chamber EED cell to remove phenol from wastewater, which cannot be easily removed by conventional ED. Compared to three-chamber EED cell, the energy consumption in a two-chamber EED is lower due to the lower resistance of the two-chamber membrane stack, however, the three-chamber EED cell can produce purer products. Therefore, when choosing and designing an EED cell, it is important to take conversion ratio and energy efficiency into consideration (Lei et al., 2020). Wei et al. (2013) compared the performance of EED and bipolar membrane electro-dialysis (BMED) in regenerating sodium hydroxide from spent caustic and they concluded that EED is less economically feasible for large scale application because each EED unit needs one pair of electrodes. Furthermore, it is reported that EED performance is strongly affected by CEMs, and cell voltage drop during EED operation is a critical problem. Therefore, in order to lower energy consumption and maintain cell voltage during an EED process, one future research direction is to optimize membrane properties to improve the current efficiency (Das et al., 2019b; Tanaka et al., 2019).

To conclude, IEMs are widely used in a variety of applications, and some critical challenges and future research directions are summarized in Table 6. Generally, these challenges include cost reduction, fouling control, stability improvement, and energy consumption reduction. Correspondingly, future research should aim to develop low cost IEMs with antifouling performance, high chemical, thermal and mechanical stabilities, and high conductivity.

## 5. Conclusion

Research interest in ion exchange membranes (IEMs) is growing rapidly in recent years. Membrane synthesis and applications are closely related, where diverse synthesis techniques enable the widespread applications of IEMs, while the diverse applications of IEMs also necessitate the synthesis of different types of IEMs to meet different requirements. Among the various synthesis methods, green synthesis of IEMs has a bright future but currently it is still at an early stage and more research is needed to explore renewable membrane materials and to develop organic solvent-free synthesis methods. For the application side, water and energy are two major applications of IEMs, where the water-based applications mainly include electro-dialysis, diffusion dialysis and membrane capacitive deionization, while the energy-based applications mainly include reverse electro-dialysis, fuel cells, redox flow battery and electrolysis. From the membrane perspective, to make these applications more effective and efficient, future research is needed to explore low-cost membrane materials, and to develop membranes with high ionic conductivity, high permselectivity, high stability and long durability. Furthermore, as different applications could have special requirements for the membrane properties, it is important to take these factors into consideration when tuning membrane properties.

**Table 6**  
Summary of current challenges and future development directions of IEM applications.

Applications	Current challenges and future development directions
Electrodialysis (ED)	(1) It is challenging to control IEM fouling during ED, especially when ED is used for high concentration industrial wastewater treatment, so future research is needed to develop IEMs with improved antifouling properties; (2) it is challenging to achieve satisfying treatment performance when the compositions of feedwater are complex, and research is needed to optimize ED design, such as coupling with other technologies to improve the overall performance.
Bipolar membrane electro dialysis (BMED)	It is challenging to maintain the stability of the two layers in the membrane, which hinders the stable operation of BMED, and future research should aim to develop more stable and durable membranes.
Electrodeionization (EDI)	It is challenging to stabilize the position of resin beads in EDI, and more research is needed to address this issue.
Diffusion dialysis (DD)	(1) DD process is very slow as the driving force is very weak, so future research is needed to improve the efficiency; (2) more research is needed to develop IEMs for DD application, e.g., to reduce the thickness and compactness of the membranes to improve acid recovery capacity and efficiency.
Membrane capacitive deionization (MCDI)	(1) MCDI is still at its early stage, and more research is needed to gain a better understanding of the process and optimize the parameters; (2) more efforts are needed to improve the energy efficiency of MCDI, e.g., by developing IEMs with high ionic conductivity and integrating them with the electrodes.
Reverse electro dialysis (RED)	Currently RED is not cost-effective and there are also some technical issues hindering the large-scale application of RED, and one future research direction is to innovatively redesign the RED stack components and system to lower operation cost and improve power output.
Proton exchange membrane fuel cell (PEMFC)	(1) Currently PEMFC is an expensive technology due to the high production cost of high purity hydrogen, and one research direction is to develop catalytic electrode materials which can function well in the presence of impurities; (2) the commonly used IEMs are not stable at high temperature PEMFC operation, so research is needed to develop low-cost membrane materials with excellent stability at high temperatures and high conductivity.
Anion exchange membrane fuel cell (AEMFC)	Durability is one of the most challenging issues hindering the commercialization of AEMFC, and one future research direction is to develop novel AEMs with desired cationic groups, high conductivity and thermostability.
Microbial fuel cells (MFCs)	(1) High cost of membranes; (2) the efficiency of membranes in channeling protons could be hindered because of biofouling, oxygen crossover and substrate loss, so one important future research is to develop low cost membranes with anti-biofouling properties.
Redox flow battery (RFB)	There are some critical issues, such as the high price of perfluorinated membranes, crossover effects, pumping losses, shunt current loss, limited energy density, degradation of the system due to the harsh environment, etc. More efforts are needed to address these issues and improve the system performance, which can be done by developing more effective and low-cost membranes that are partially fluorinated and non-fluorinated, with high ionic conductivity and low vanadium ion permeability to achieve high efficiency.
Electrolysis for H <sub>2</sub> production	

**Table 6 (continued)**

Applications	Current challenges and future development directions
Electrolysis for chlor-alkali industry	(1) Currently electrolysis for H <sub>2</sub> production is not economically favourable due to its high operational cost, so future research is needed to address this issue; (2) expensive catalytic electrode materials (e.g., platinum) are used in PEM-based electrolysis, so future research should aim to develop cost-effective catalyst alternatives; (3) for AEM-based electrolysis, more efforts are needed to develop membranes with high conductivities, good mechanical strength, and especially, high stability in alkaline conditions.
Electro-electrodialysis (EED)	(1) Chlor-alkali process is very energy-consuming and costly, so future research is needed to address this issue, e.g., by developing highly active electrocatalysts; (2) more research is needed to prolong the lifetime of IEMs, e.g., by improving membrane durability in alkaline environment and resistance to chemical attack.
	Energy consumption in the EED process is high, and cell voltage drop is a serious challenge, so future research is needed to reduce energy consumption and maintain cell voltage, e.g., by optimizing CEM properties.

### 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.

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### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.chemosphere.2021.130817>.

### Abbreviations

AEL	Anion exchange layer
AEMFC	Anion exchange membrane fuel cell
AEM	Anion exchange membrane
AER	Anion exchange resin
BMED	Bipolar membrane electro dialysis
CDI	Capacitive deionization
CEL	Cation exchange layer
CEM	Cation exchange membrane
CER	Cation exchange resin
CMME	Chloromethyl methyl ether
DD	Diffusion dialysis
DMAc	Dimethylacetamide
DMF	Dimethylformamide
DMSO	Dimethyl sulfoxide
ED	Electrodialysis
EDI	Electrodeionization
EDR	Electrodialysis reversal
IEC	Ion exchange capacity
IER	Ion exchange resin
IEM	Ion exchange membrane
MCDI	Membrane capacitive deionization
MFC	Microbial fuel cell

MMM	Mixed matrix membrane
NaSS	Sodium 4-vinylbenzenesulfonate or sodium styrenesulfonate
NIPS	Non-solvent induced phase separation
NMP	N-methyl-2-pyrrolidone
PBI	Polybenzimidazole
PEEK	Polyether ether ketone
PEMFC	Proton exchange membrane fuel cell
PEM	Proton exchange membrane
PES	Polyether sulfone
PPO	Polydimethyl phenylene oxide
PSS	Polystyrene sulfonate or sulfonated polystyrene
PTFE	Polytetrafluoroethylene
PVA	Polyvinyl alcohol
PVC	Polyvinyl chloride
PVDF	Polyvinylidene fluoride
RED	Reverse electrodialysis
RFB	Redox flow battery
RO	Reverse osmosis
SCIE	Science Citation Index Expanded
SPBI	Sulfonated polybenzimidazole
SPEEK	Sulfonated poly(ether ether ketone)
SPES	Sulfonated polyethersulfone
THF	Tetrahydrofuran
TIPS	Thermally induced phase separation
VIPS	Vapor-induced phase separation
VRFB	Vanadium redox flow battery

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