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Desalination and Water Treatment www.deswater.com doi: 10.5004/dwt.2021.26729 213 (2021) 214–228 February Current progress in metal-organic frameworks-embedded membranes for water desalination Bary Leonard Suwandia,b, Laurensia Nadya Widjajaa,b, Stefani Catherinea,b, Shella Permatasari Santosoa,b,*, Felycia Edi Soetaredjoa,b, Artik Elisa Angkawijayac, Adriana Anteng Anggorowatia, Suryadi Ismadjia,b,*, Phuong Lan Tran-Nguyend, Yi-Hsu Jub,c aDepartment of Chemical Engineering, Widya Mandala Surabaya Catholic University, Kalijudan 37, East Java, Surabaya 60114, Indonesia, Tel. +62 31 3891264; Fax: +62 31 3891267; emails: shella_p5@yahoo.com (S.P. Santoso), suryadiismadji@yahoo.com (S. Ismadji), baryleonard10@gmail.com (B.L. Suwandi), laurensianadya@gmail.com (L.N. Widjaja), stefanicatherine99@gmail.com (S. Catherine), felyciae@yahoo.com (F.E. Soetaredjo), adrianaanteng@ukwms.ac.id (A.A. Anggorowati) bDepartment of Chemical Engineering, National Taiwan University of Science and Technology, 43 Keelung Rd. Sec. 4, Taipei 10607, Taiwan, email: yhju@mail.ntust.edu.tw (Y.H. Ju) cGraduate Institute of Applied Science and Technology, National Taiwan University of Science and Technology, 43 Keelung Rd. Sec. 4, Taipei 10607, Taiwan, email: artikelisa@mail.ntust.edu.tw (A.E. Angkawijaya) dDepartment of Mechanical Engineering, Can Tho University, 3-2 Street, Cantho City, Vietnam, email: tnplan@ctu.edu.vn (P.L. Tran-Nguyen) Received 7 May 2020; Accepted 23 October 2020 abstract Rapid population and economic growth cause severe anthropogenic emissions, which lead to many water pollutions. As a result, the demand for potable water is increasing dramatically in the last decades. Various logical solutions and sustainable developments, such as converting seawater into freshwater, have been explored. Several methods have been employed in water desalination, such as pressure-driven membranes, capacitive deionization, and adsorption desalination. Still, the lim- itations of the present technologies are low water productivity and quality. Therefore, it is essential to perform further research to design materials and methods with higher salt rejection property and large flux capacity. Recent study and development have pointed out an advanced material called metal-organic frameworks (MOFs) with fascinating chemical and physical properties. Researchers and engineers have been studying MOFs for their potential application in water desalination. This review is aimed to stimulate

future research related to the utilization of MOFs for seawater desalination, particularly in the desalination membrane development. Recent advances from the MOF in water desalination are given, starting with outlining various studies of membrane-modified MOF, followed by the application of MOF in desalination technology. Finally, the explanation of the concept of increasing desalination performance by MOF to fill the knowledge gap. Keywords: Membrane; Nanomaterial; Salt rejection; MOF; Desalination * Corresponding authors. 1944-3994/1944-3986 © 2021 Desalination Publications. All rights reserved. B.L. Suwandi et al. / Desalination and Water Treatment 213 (2021) 214–228 1. Introduction The availability of clean and freshwater is the fore- most crucial resource for all life forms on earth. The need for decent water increases with rapid population growth. Ironically, the availability of clean water has become scarce

19**due to the rapid industrialization and urbanization** (source **of**

anthropogenic emission). A severe level of emissions caused by anthropogenic activities causes drastic deple- tion of clean water sources [1]. At present, there are more than 0.78 billion individuals around the globe who do not have access to clean water resources, which have resulted in significant health issues. Reliable access to clean and affordable water is viewed as one of the global problems that require special attention in the 21st century; the sus- tainable development of water desalination and purifica- tion has become a routine agenda for many engineers to overcome the issue. Desalination (Fig. 1) is considered as a sustainable solution in providing freshwater supplies. Attempts to desalinate seawater by using methods such as forward osmosis, reverse osmosis, evaporation, capacitive deionization, adsorption, and distillation to gain freshwater has been made to fulfill this increasing demand [2–6]. At present, reverse osmosis membrane technology has become the most utilized desalination process due to its potenti- ality to diminish the process cost, without degrading the water quality. Numerous studies on the desalination process using membrane have been conducted to increase the efficiency of obtaining fresh water from the sea or brackish water. In recent years, nanotechnology and nanomaterials gradu- ally play a vital role in improving desalination technology [7,8]. Membrane constructed from nanomaterials is consid- ered one of the best filtration-materials in desalination [9]. Nanomaterials are described as the material smaller than 100 nm in at least one external dimension; its small size pro- vides a higher surface area to volume ratio [10]. Reviews about nanomaterials

19**for water and wastewater treatment** in various applications **such as**

photocatalysis, water puri- fication, adsorption, disinfection, sensing, and monitor- ing have been written, as summarized in Table 1 [10–15]. The modification of thin-film membranes with various nanomaterials gives us a chance to redefine the standard of water desalination processes. The blossoming research about water desalination using nanomaterials left us with many choices of process Fig. 1. Schematic diagram of saline water desalination process. and many choices of material, as summarized in Table 2. A clear insight into the challenges and possibilities of water desalination is the key step to develop better tech- nology for fulfilling the increasing demand for freshwater at a rationally low cost. Lately, the utilization of metal-or- ganic frameworks (MOFs) as new membrane materials in water desalination is gaining much interest among engi- neers. The enormous pore of MOFs provides a large sur- face area, making them efficient in water desalination. This review aims to explore and summarize the current development of MOF-embedded membranes and their disadvantages and possible future advances, which yet to be done. This review will provide insight to the reader in seeing the promising role of MOFs in increasing the effi- ciency of desalination and its current progress. Also, it is noted that membrane modification with MOFs is an attrac- tive approach to improve the membrane properties, which resulted in higher salt rejection and water permeability. We foresee the rapid development of MOFs utilization in dif- ferent desalination processes and how the incorporation of MOFs can improve membrane performance. Modification of desalination membranes by embedding MOF has been widely studied, where the addition of MOF provides a notable

increase in desalination performance. For example, the addition of MOF can reduce the affinity between water molecules and the pore surface of the mem- brane, making it easy for water molecules to pass through the pores. Others, the addition of MOF, brings the pres- ence of antifouling or antibacterial properties to the mem- brane. This article discusses the performance of several MOF-modified membranes, their preparation, and how MOF can improve their desalination performance. 2. Assorted MOF-modified membranes Based on the size of the materials, the membranes cat- egorize as thin-film nanocomposite (TFN) and thin-film composite (TFC). TFN is the membrane-embedded by nanoparticles, while TFC is embedded with larger particles. Polyamide (PA) thin-film is the most common membrane used in desalination; its fabrication involves the interfacial polymerization (IP) process in

8**which m-phenylenediamine (MPD) and trimesoyl chloride (TMC**

) are reacted [29]. In this section, various type of MOF-embedded membranes is presented. 2.1. Simple MOF-embedded membranes The incorporation of silver-based MOF, namely Ag-BTC (Ag = silver(I) and BTC =

4**1,3,5-benzene tricarboxylic acid), into the polyamide**

(PA) thin-film was reported to provide a significant increase in

21**desalination performance. The membrane was** prepared **by**

using the IP method, a differ- ent concentration of AgBTC was loaded in an organic solu- tion

21**containing 14 wt.% polyethersulfone (PES) and 1 wt.% polyvinylpyrrolidone (PVP) dissolved in DMF. The**

result- ing MOFs, with average diameters of 33.4 nm (nanocrys- tals), were then embedded into the membrane at various weight percentages. It is reported that TFN-0.04 and -0.08 (TFN membrane with 0.04 and 0.08 wt.% AgBTC loading) was able to enhance the membrane water permeability 1.3 216 B.L. Suwandi et al. / Desalination and Water Treatment 213 (2021) 214–228 Table 1 Progress on water desalination using nanomaterials Nanomaterials Emphasis of review References Nanomaterials-polymer composite Incorporation of carbon-based nanomaterials (CNTs) into polymer composite membranes membranes to form membranes with exceptionally high salt-rejection and permeability. Metal-oxide nanoparticle- Development of the coating of hydrophobic membranes with metal-oxide enhanced membranes, nanoparticles and carbonaceous nanomaterials such as CNT for various CNT-modified metal-oxide applications, including for desalination process. The functionalization nanoparticles of the membrane into hydrophilic to impart its mechanical properties was also discussed. Enhanced thin-film composite Incorporation of nanoparticle into a thin-film composite membrane to enhance membranes (TFC) the low-biofouling and anti-scaling properties of the TFC membrane. The zeolite-embedded membranes exhibit exceptional salt-rejection activity. Zeolites, Aquaporin The use of zeolite and aquaporin for the desalination process. Ion exchange membranes (IEMs) Developments of IEMs include the utilization of nanomaterials such as CNTs, zeolites, silica, TiO2, and silver nanoparticles to enhance the strength, permeate-selectivity, ion-conductivity, and thermal stability of the membrane. Rationally fabricated The use of specific nanomaterials (graphene 2-D, zeolite, and molybdenum nanomaterials disulfide) for the production of 3-D nanomembrane for application in a solar desalination device. Graphene The use of graphene as

nanofiltration materials. The improvement of the membrane structure to enhance the water flux was also discussed in this review. Carbon nanotubes, zeolites, The use of CNTs and zeolite membranes for seawater desalination. Some critical features of CNTs and zeolite as desalination membranes were also discussed. Aquaporin membranes, Utilization of carbon-based materials (CBM), and aquaporin as promising Carbon-based membranes membrane materials. Various aspects of the desalination process using these membranes are also discussed. Nano-fiber membranes The use of nanofiber membranes for removing salt in the desalination process. Zeolites, CNT Various aspects of the application of zeolites and CNT in desalination and water treatment process. Nanoparticle oxides, carbon The synthesize of capacitive deionization electrodes based on electrosorption nanotubes, graphene, carbon nanomaterial electrodes for desalination. A comparison with reverse nanofibers, and nonporous osmosis is also given in this review paper. carbon cloth CNT membranes with Modification of CNT membranes with antimicrobial nanoparticle for antimicrobial nanoparticles the water desalination process. (silver nanoparticle and TiO2) Graphene-based membrane, Modification of carbon nanotubes and graphene by chemical treatment carbon nanotubes for the desalination process. Carbon nanotube Application of carbon nanotubes (CNTs) as high selectivity desalination membranes. Graphene-based nanomaterials Application of graphene-based nanomaterials as an innovative solution for water desalination. CNT, inorganic membrane Production and application of CNT based and inorganic membranes for the desalination process. Metal-organic framework (MOFs) The desalination process depends

> 1**on the** MOFs' **pore size**, where **water molecules** are possible **to pass, but salt ions** do not pass. **The**

absence of cooperation between water flux and salt rejection depends on MOFs' pore size and surface charges. [9] [16] [3] [1] [17] [18] [19] [20] [21] [22] [23] [24] [2] [25] [26] [27] [28] [29] T able 2 Performance of some hybrid membrane in different desalination processes Desalination process RO RO RO RO RO RO FO FO FO FO MOF-layers ZIF-8/P A MOF1/PES/P AN MOF2/PES/P AN UiO-66/PSU MIL-125/PSU POSS/P A with POSS = • P-8Phenyl • P-8NH3Cl • P-8NH2 • P-1NH2 UiO-66/Al hollow fiber HKUST -1@P A/PES Ag-BTC/PES/PVP CuBDC-NS/P A T est conditions p: 15.5 bar C: 2,000 ppm NaCl v: 0.37 m/s p: 5 bar C: 7,000 ppm NaCl p: 300 psi C: 2,000 ppm NaCl p: 15.5 bar C: 2,000 ppm NaCl v: 0.37 m/s p: 10 bar C: 0.20 wt.% (of the salt used) p: 2 or 4 bar C: 500/2,000 ppm NaCl C: 0.5 – 2 M NaCl v: 21 cm/s C: 1 M NaCl v: 15 cm/s PMMF300 C: 0.5 M MgCl2 (draw solution) PMMA100 PMMC300 (PMM prepared from mix- ture of substrate and active layer(AL)) ZnO@MOF-5 (magnetic MOF-5) C: 0.5 M Caspian seawater W ater permeability (L/m2 h bar) 3.35 36 (5 bar) 41 (5 bar) 74.9 (300 psi) 85.0 (300 psi) 33.0 (15.5 bar) 27.1 (15.5 bar) 33.4 (15.5 bar) 3.2 (15.5 bar) 0.28 6.94 ~4.3‡ 3.13 ~92 (at given p)‡ ~98 (at given p)‡ ~107 (at given p)‡ 117 (at given p) Solute rejection (%) or permeability >98% 93% 97% 98.8% 98.6% 98.2% 98.9% 98.6% 95.9% 86.3% (Ca2+) 98.0% (Mg2+) 99.3% (Al3+) ~98.2% (2 bar) ~97.4% (4 bar) ~0.6 L/m2 h 0.317 L/m2 h ~75%‡ ~91%‡ ~93%‡ – Ref. [30] [31] [32] [33] [34] [35] [36] [37] [38] B.L. Suwandi et al. / Desalination and Water Treatment 213 (2021) 214–228 [39] 217 218 Cross-flow filtration CDI Electrochemical cell CDI Electrochemical cell Adsorption Adsorption MD MD Ultrafiltration MD ultrafiltration MD MD ultrafiltration NH2-MIL-101(Al)/chitosan ZIF-67/PPy PC-ZnCo-3 CPO-27(Ni)ζ Aluminum fumarateζ MIL-101(Cr)ζ MIL-101(Cr)/CaCl2ζ AlFu/PVDF CA/HKUST -1@GO TMU-5/PES Si NPs/PVDF hZIF-8/PSU C: 2,000 mg/L NaCl, 1,000 mg/L MgCl2 or CaCl2 C: ~10 mM NaCl v: 10 mV/s† V: 1.2 Volt C: 750 mg/L NaCl V: 1.4 Volt Thot: 150°C Tcold: 25°C Cycle: 700 s Switching: 70 s – C: 3.5 wt.% NaCl p: 0.15 MPa C: 1.0 g/L BSA p: 3 bar C: 40,000 ppm oil-water emulsion p: 1.2 kPa C: 35 g/L NaCl p: 0.2 MPa C: 0.5 g/L BSA 4.0 (NaCl) ~3.8 (MgCl2)‡ ~3.4 (CaCl2)‡ – – – – – 183.51 (0.15 MPa) 123.212 (kg/m2 h) 12,749.6 (g/m2 h) 579 (0.2 MPa) ~29.0% (NaCl)‡ 93.0% (MgCl2) 86.5% (CaCl2) 11.34 g salt/g (with 1.32)δ 45.62 mg salt/gδ

> 1**0.47 gH2O**/gδ **0.53 gH2O**/gδ **1.47 gH2O**

/gδ 0.65 gH2O/gδ 99.9% 95.37% >98% ~100% >98% [40] [41] [42] [43] [44] [45] [46] [47] [48] [49] †Scan rate in millivolt per second; ‡Value approximated from the given graph in cited literature; δDesalination or adsorption capacity; ζNon-membrane; PA = polyamide; PES = polyethersulfonate; PAN = polyacrylonitrile; PVP = polyvinylpyrrolidone; PSU = polysulfone; PPy = polypyrrole; PC = porous carbon; CA = cellulose acetate; BSA = bovine serum albumin; PVDF = polyvinylidene fluoride. MOF =

> 4**metal-organic framework; ZIF = zeolitic imidazolate framework**

; BTC = trimesic acid (

> 4**1,3,5-benzene tricarboxylic acid**

); BDC = benzene-1,4-dicarboxylic acid; POSS = polyhedral oligomeric silsesquioxane; UiO = University of Oslo (prototypical of Zr-MOF); MIL = Materials Institute Lavoisier; A100 =

> 12**Basolite A100 (MIL 53 or** aluminum **terephthalate**); C300 = **Basolite C300 (HKUST-1 or copper benzene-1,3,5-tricarboxylate**); F300 = **Basolite F300 (Fe-BTC**

); AlFu = aluminum fumarate; Si NPs = silica nanoparticles; hZIF = tannic acid modified ZIF. B.L. Suwandi

> 13**et al. / Desalination and Water Treatment** 213 (**2021**) 214–228 **B**

.L. Suwandi

> 13**et al. / Desalination and Water Treatment** 213 (**2021**) 214–228 **Fig. 2**. Schematic representation **of**

PMM compared to traditional MMM (redrawn with slight modification from Lee et al. [6]). and 2.4 times higher, compared to the polyamide mem- brane. The MOF loading also led to the enhancement of seawater desalination flux, 34 L/m2 h for TFN-0.04, while only 27 L/m2 h for TFC [36]. UiO-66 and MIL-125 are other nano-sized MOFs that have been proven to have excellent desalination performance. Significant water permeabil- ity of

> 20**74.9 L/m2 h, with** a high salt **rejection** of **98**

.8%, was achieved by modifying the

> 17**TFN membrane with UiO-66**. The modification of **the**

TFN membrane with MIL-125 pro- vided higher surface area and pore size. Thereby, higher water permeability of 85

24**L/m2 h and** 98.6% **salt rejection were obtained**

[29,32]. Desalination performance of binary and ternary MOFs was reported; namely, MOF1 synthesized from the triva- lent cerium cluster in a warm solution of carboxymethyl cellulose (CMC); and ternary complex, MOF2, which is synthesized by the addition of anthranilic acid to MOF1. The implantation of cerium-based MOF (0.1 wt.%) into the TFC can increase NaCl rejection from 69% to 93% (for MOF1) and 97% (for MOF2), from an initial of 7,000 ppm NaCl at a feed pressure of 5 bar. Higher MOF loading helps to increase the water flux 1.2 times from TFC only [31]. Dai et al. [37] embedded

4**copper 1,4-benzenedicarboxylate nanosheets (CuBDC-NS**, BDC = benzene-1,**4**

-dicarboxylic acid) inside the PA layer. This study shows that CuBDC-NS embedded inside the PA layer helps in increasing the water flux by nearly 50% (from 1.95 to 3.13 L/m2 h bar), and decreasing salt permeability by 25% (

17**from 0**.418 **to 0**.317 **L/ m2 h), compared to the** plain **membrane**

. The addition of CuBDC-NS also resulting in the enhancement of antifoul- ing activity due to its hydrophilicity and biocidal ability. While almost all of the studies used hydrophilic MOFs, Duan et al. [33] introduced a unique approach in utilizing the TFN membrane for water desalination. MOF with hydrophobic properties was used instead of hydro- philic MOFs, specifically the ZIF-8 (ZIF = zeolitic imidaz- olate framework). The addition of 0.4% (w/v) nano-ZIF-8 (~200 nm) to TFN membrane increases NaCl rejection up to 62%, with water permeability of 3.35 L/m2 h bar (higher than BW30). A similar approach has been demon- strated by other researchers, such as hydrophobic-POSS (POSS = polyhedral oligomeric silsesquioxane) implan- tation to TFN membranes produces a membrane with rigid and bulky cages which effective in increasing the volume of selective membrane layers [33]; incorporation of porous-hydrophobic carbon nanotubes into TFN produces a membrane with ultra-fast water transport [50]. 2.2. Multiple MOFs-embedded membranes Several MOFs can be combined in desalination mem- branes, such as in preparing mixed-matrix membranes (MMMs). MMMs are a novel material that gains a lot of attention due to their superior thermal stability and mechanical properties. The synthesis of MMMs

12**involves (1) dispersion of MOF**-nanoparticles into **organic solvent; (2**) dissolving **polymer**

materials into the MOF solution with constant stirring to produce a homogeneous mixture; (3) evaporation of water in the casting of the solution. MOF- based MMMs are highly beneficial in the water desalination process due to their tunable and small pore size, which come in handy for adjusting water permeability [38]. An MMM membrane prepares by casting two different MOFs, namely

5**NH2-MIL-101(Al) and NH2-MIL-101(Cr**), onto **a**

polysulfone (PSU) support sheet. The resulting TFN membrane has a positively charged surface that allows for a higher rejection of multivalent positive ions. In contrast, monovalent positive ions are less rejected—this

caused a vast rejection of salt, such as MgCl2 and CaCl2 (93.0% and 86.5%, respectively), whereas the rejection of NaCl and Na2SO4 fell drastically. The electrostatic effect makes the

5**salt rejection order to be MgCl2 > CaCl2 > NaCl > Na2SO4**

[40]. Wang et al. [51] report another notable MMM; however, clay minerals were used instead of MOFs. The membrane is prepared by using polyvinylidene fluoride (PVDF)/cloisite

15**clay composite hollow fibers with the addition of**

ethylene glycol (EG) and N-methyl-1-pyrrolidone (NMP). The result- ing membrane has a low heat conductivity, low vapor trans- fer, high thermal efficiency, and stable mechanical strength. The membrane also has proven to enhance the permeate flux in the MD process, with 100% salt rejection [51]. 2.3. Porous MOF-embedded membranes Unlike MOFs-based MMMs, the porous matrix mem- branes (PMMs) use water-soluble MOFs nanoparticles as filler, which will be easily washed away by water. The idea is to provide more holes on the membranes, to improve the flux of solvent through the membrane [29]. Fig. 2 shows the difference between MMM and PMM. PMM filled with MOF was developed by Lee et al. [38], it has been used for forward-osmosis processes. In the fabrication of MOFs- based PMM, Lee et al. [38] mixed three different types of MOFs, that is Basolite F300, Basolite C300, and Basolite A100. Polyacrylonitrile (PAN) was used as the dope solution, and B.L. Suwandi et al. / Desalination and Water Treatment 213 (2021) 214–228 the addition of lithium chloride into the dope solution was processes. Ions from saline water are removed by apply- conducted at 60°C. The layer-by-layer (LbL) self-assembly ing electrostatic forces onto the solution through positively process was done by immersing MOFs into polyallylamine and negatively charged electrodes. Cations and anions hydrochloride (PAH) and polysodium 4-styrene-sulfonate are attracted to their corresponding electrodes, Fig. 3a. (PSS) for three times to form the rejection layer. Finally, it The regeneration step is also necessary to desorb the ions was immersed in 0.1 wt.% glutaraldehyde (GA) for 30 min from saturated electrodes, which then transferred into to establish the crosslinking of the salt rejection layer, which wash water by reversing the potential difference between provides high forward osmosis water flux. PMM can alto- electrodes, as shown in Fig. 3b [24]. gether increase water penetrability in a pressure-driven The improvement on CDI process can be engaged by membrane process, perhaps due to the expulsion of MOF changing the properties of the electrodes, such as into (1) particles in the polymer matrix, which expands membrane high surface region that can be accessed by large-sized porosity and interconnectivity. More critically, current ions, (2) high electronic conductivity, and (3) high elec- researches recommend MOF as a green layout might have trochemical stability [54]. The current interest in research great potential for pressure-driven membrane processes as from membrane CDI (MCDI) is focused on structuring new well as for carbonated nanofiber membranes for particular materials with alluring properties, as referred above [42]. screening and separation of nanoparticles [52], or osmoti- The main idea was to combine the high porosity MOFs and cally driven membrane processes (i.e., osmosis to the front high conductive electrode to produce a newly effective elec- (FO) and pressure-suppressed osmosis (PRO)) [53]. trode. One of the latest attempts on developing high-end electrodes for the CDI system was conducted by Wang et al. [41]. A combination of ZIF-67 with polypyrrole (PPy) 3. MOFs-modified membrane in various nanotubes resulted in a three-dimensional hybrid mate- desalination methods rial with interconnected MOF particles using the nano- 3.1. Capacitive deionization tubes as bridges; the bridge enables the MOF particles to transfer electrons between them. The well-dispersed MOF Capacitive deionization (CDI) was first aroused during particles gave uniform particle size distribution on the the

9**1960s and has** multiplied **over the past two decades as**

hybrid electrode. This ZIF-67/PPy hybrid electrode exhib- a promising water desalination innovation [54]. The idea ited a remarkable desalination capacity of 11.34 mg/g and of CDI technology is to temporarily storing salt ions in an excellent cycling stability [41]. A series of bimetal MOFs electric double layer (EDL) at the interface between the CDI (BMOFs) with various Zn to Co molar ratios were suc- electrode and the salt solution. This technology offers low cessfully synthesized to design the optimal combination energy consumption because the voltage utilized is typi- of ZIF-8 and ZIF-67, which later be used to create a CDI cally

> 9**lower than the electrolysis** potential **of water (~1.23 V**

) electrode. The recently arranged BMOFs were coded as [55]. The CDI requires both deionization and capacitive BMOF-ZnCo-n, and the derived carbon was characterized (a) (b) Fig. 3. (a) Deionization of saline water and (b) regeneration of CDI electrodes (redrawn with slight modification from Gaikwad and Balomajumder [24]). B.L. Suwandi et al. / Desalination and Water Treatment 213 (2021) 214–228 as PC-ZnCo-n (n = molar proportion of Zn to Co). Among the synthesized PC-ZnCo-n,

> 9**PC-ZnCo-3** demonstrated **the highest salt removal capacity of 45.62 mg/g in** the membrane CDI **system**

[42]. 3.2. Forward osmosis Among the

> 2**methods applied** for **water and wastewater treatment, FO** provides **an**

option in contrast to conven- tional pressure-driven membrane processes (particularly in seawater desalination); that is, minimal cost, high selec- tivity, and less inclination to fouling contrasted with other membrane methods [56,57].

> 2**Membranes with two** unique **structures are** utilized **in** the **FO** procedure: (**i**) robust **skin** asymmetrical **membranes containing** layers of **skin**

coor- dinated over the permeable layer [58], and (

> 2**ii) thin-film composite (TFC) membranes, which** comprise **of ultra**-slim **active** layers (**AL**

) over the permeable support layer (SL) [59]. Fig. 4 shows the FO process. Briefly, the water mole- cules pass through the membrane

> 14**from the feed** water, **which has a** lower concentration **to the draw solution**

and higher concentration due to osmotic pressure. The mem- branes utilized in the FO are mainly adopted in a flat sheet configuration [60]. However, most of the flat sheet config- urations fold into spiral wounds that

form tubular mem- branes. In maximizing the surface area of the membrane, the configuration of the tubular membrane or hollow fiber must be used. An ongoing research effort concentrated on building a

> 14**high-performance hollow fiber** membrane **for** the **FO**

procedure [61]. In

> 14**large-scale FO** processes, **hollow fiber membranes** show **more advantages than** that of **flat sheet membranes**

. Hollow fiber membranes offer an increase in durability, flow patterns, and packaging density [62]. Lee et al. [38] utilized PMM as SL

> 4**for the fabrication of TFC FO membranes. The** bulk porosity **of**

MOF-based PMM expanded as the membrane tortuosity decreased— this led to the increased

> 2**mass transfer of water and the well-controlled**

dilutive internal concentration polar- ization (DICP) in the FO substrate due to the decreasing membrane fundamental parameter [38]. Another effort for developing this research was made by Arjmandi et al. [39]; a

> 2**magnetic water-unstable MOFs** was used **to** create **pores with** various sizes **and**

dispersions throughout the membrane. Zirehpour et al. [36] developed hydrophilic silver-based MOF-embedded TFC

> 17**to enhance the** desali- nation **performance of the** FO **membrane. The**

prepared membranes have an improved porosity and pore intercon- nectivity, which resulted in a high FO flux of 82 L/m2 h. Furthermore, the hydrophilic surface and high porosity of MOF provide another transport way for water [36]. 3.3. Reverse osmosis The

> 20**most commonly used** water **desalination process is**

RO, with

> 1**around 20,000 large scale plants worldwide. The** concept of RO **is the**

inverse of FO; by giving a more massive pressure

24**than the** salinity **osmotic pressure of the feed** water, **the**

water molecules are forced

1**to pass through a** set of **semi-permeable** membranes and leaving **the salt ions** behind (**Fig**

. 5). The main advantages of the RO pro- cess are high energy efficiency, low operating temperature, and increased productivity [29]. The HKUST-1 MOF has been used to modify the PA matrix with PES and polydo- pamine (PDA) SL, in a low-pressure RO process. HKUST-1 considers as the potential material because of its fast trans- port of water molecules, which pass through the flow chan- nels of the organic framework, and highly solute rejec- tion due to its steric exclusion properties [35]. The formed membrane resulted in the high rejection of NaCl, which was 98.2% at 2 bar and 97.4% at 4 bar. For a long term RO operation, the modified HKUST-1 at the PA membrane shows the same durability as the original membrane. The HKUST-1 doped membrane has enhanced surface hydro- philicity, antifouling capability, and water permeability compared to the original membrane. The higher surface hydrophilicity allows the water molecules to be adsorbed into the PA matrix, facilitating the water transport through the porous flow channels. Moreover, the negative sur- face charges lead to higher fouling resistance [30,33,35]. 3.4. Adsorption desalination AD is an emerging alternative for the water desalination process due to its environmentally friendly, low-cost, and high energy efficiency properties. AD technology mainly consists of two cycles, that is evaporation–adsorption and Fig. 4. Forward osmosis schematic diagram. B.L. Suwandi et al. / Desalination and Water Treatment 213 (2021) 214–228 Fig. 5. Reverse osmosis schematic diagram. desorption–condensation. The first cycle works by evapo- rating water using a low-pressure evaporator. Adsorbent materials then collect the water vapor in two or more beds containing a finned tube heat exchanger. After the evapo- ration step, hot water is employed through the heat-ex- changer tube to warm the beds so that the water vapor release. Cooling water is supplied afterward into a con- denser above the beds, which condense the vapor into collectible potable water [29,43]. The system consists of two adsorption beds packed with MOF, evaporator, and condenser material, as shown in Fig. 6. AD system with a silica gel bed is the most common one. However, the major disadvantage is the lack of hydro- philicity of this material that causes a low adsorption capac- ity, which must be covered up by employing high relative pressure. The utilization of porous and highly-hydrophilic MOFs has been an exciting way to cope with the problem. The improvement of AD by incorporation of MOF has been demonstrated by Elsayed et al. [43], in which they are employing three kinds of MOF materials, namely MIL- 101(Cr), aluminum fumarate, and CPO-27(Ni). MIL-101(Cr)- employed bed exhibited the highest water uptake and also the highest specific daily water production (SDWP), which outperformed the other two MOFs. Fig. 6. Adsorption system schematic diagram (Redrawn with slight modification from Elsayed et al. [43]). 3.5. Membrane distillation MD has been developed for over 40 y, and it is con- sidered a potential application for seawater desalina- tion due to its high productivity and low-cost operation. In particular, membrane distillation involves water evap- oration and vapor condensation and mass and heat trans- fer mechanisms, as shown in Fig. 7. A porous hydrophobic membrane was installed as thermal insulation and physical barrier between the two phases; the membrane allows

15**free transport of water vapor through the pores** while prevent- ing **the liquid** water **from getting** off **the membrane due to** its **surface tension forces**

.

22**In the MD process, the membrane**

must be kept dry so that only vapor diffuse

22**through the pores** [41]. **The hydrophobic membrane**

mostly comprises of

23**polyvinylidene fluoride (PVDF), polytetrafluoroethylene (PTFE), polyethylene (PE), or polypropylene (PP**

); which Fig. 7. Membrane distillation (MD) module basic setting (Redrawn with slight modification from Mahmoudi and Akbarzadeh [63]). B.L. Suwandi et al. / Desalination and Water Treatment 213 (2021) 214–228 have the following properties: high liquid entry pressure, high permeability, low fouling capability, high chemical stability, and high thermal stability [63–65]. The unique properties of the MD process rely on its thermal gradient, which is

11**created across a hydrophobic membrane and** func- tionates **to** bring **the** solution **to their saturation point** with- out **any** noteworthy **flux-decline. The** membrane used in **the** MD **process**

holds all solid or nonvolatile contaminants and therefore resulting in 100% pure water. A green modi- fication of the MD process has been developed, particularly for its energy source; the

11**solar energy, geothermal energy, and waste grade energy (with low-temperature industrial streams**

) has been applied to MD technology [65]. Many engineers have studied sustainable development for membrane used in MD technology, where the incor- poration of MOF to synthesis a hybrid membrane is the most popular to date. The main goal of the development is to create a membrane with high porosity to achieve an efficient (both thermally and permeability) water desali- nation. Cheng et al. [45] synthesized a

23**novel hydrophobic hybrid MOF/PVDF hollow fiber** membrane (**MOF**

= alu- minum fumarate, abbreviated as AlFu). The hydrophobic hybrid membrane was found to improve the permeate flux and thermal efficiency because of the enlarged porosity of the membrane. The prepared MOF/PVDF shows a high salt rejection of 99.9% for 3.5 wt.% NaCl solution, during 50 h MD process. Yang et al. [66] have reported a similar finding; they have proven that incorporating iron 1,3,5-ben- zenetricarboxylate MOF into the PVDF membrane led to pore and porosity enlargement of the membrane, and subsequently improved the water flux. Incorporation of graphene oxide-modified MOF, namely HKUST-1@GO, into the membrane is reported to produce a hybrid membrane with a larger pore and smoother surface (compare to an ace- tate cellulose membrane); the HKUST-1@GO-blended mem- brane is proven to accelerate the solvent and non-solvent exchange in the PI process [66]. Other research groups also

demonstrated that a MMM modified with ZIF-8 or TMU-5 MOF has a larger pore than the pristine membrane [47,49]. The MOF (or other similar NPs) acts as nucleation materials, which cause enlargement of membrane pores post-penetration, during the membrane modification pro- cess. Incorporation of MOF produces a thermodynami- cally unstable membrane which is more active to bind the guest molecules (i.e., salts, volatile compound, and charged ions); this behavior is leading to the improvement of sol- vent and non-solvent exchange rate during the PI process. One thing to note is that excessive MOF loading into the membrane will lead to a sudden kinetic hindrance, which demoted the efficiency of solvent exchange performance. This is because a higher MOF loading will cause particle aggregation and a decrease in crystallinity [47–49,67,68]. 4. Concept of MOF-embedded membranes 4.1. Factors affecting desalination performance It has been demonstrated that the hybridization of TFN or TFC membrane with MOF can produce layers that are better compatible with the polyamide matrix, faster water transport theoretically in the framework, and hydrothermally more stable. Also, the enormous porosity of the MOFs provides a waterway for the rapid passage of water molecules through TFN or TFC layers [30,33,34]. Based on several reports, the following factors have a profound influence on the desalination process: the amount of MOF, surface area, hydrophobicity, surface charge, and inlet tem- perature. The next paragraphs outline the evidence regard- ing these influential factors. The incorporation of MOF particles in the proper amount provides an excellent contribution to improv- ing desalination performance. Increasing the number of MOFs, however, may reduce desalination performance. The addition of 0.08 wt.% AgBTC to PA thin-film, in the preparation of TFN-0.08, produced TFN with a higher B/A permeability ratio (B = solute and A = water) compare to TFN-0.04 – which means the solute permeability in TFN- 0.08 is higher than TFN-0.04. The proper addition of MOF can increase the hydrophilicity of the membrane, thus attracting the water molecules [36]. However, incorpora- tion of an excess number of MOFs is resulting in lower salt rejection (higher B/A permeability ratio) as it will crack the TFN membrane [29,32]. The higher surface area is another increase resulting from the addition of MOF to the membrane. This is since the presence of MOF in the membrane matrix causes higher pore formation, thus allows faster water permeance. Furthermore, rapid water permeation can be achieved by changing the affinity of the membrane toward the water. The addition of hydrophobic MOF (not hydrophilic MOF) results in a membrane with rapid water permeation caused by the absence of friction. The addition of hydrophobic MOF facilitates water permeability by reducing the affin- ity

5**between water molecules and the** pore **walls of the**

membrane [30,31]. Ma et al. [40] show a more intrinsic factor affecting the desalination performance, which is the surface charge of the membrane. A measurement on the surface charge of MMM, which was modified with mixed MOF NH2-MIL- 101(Al) and NH2-MIL-101(Cr), shows positive charges. The positive charge of the membrane arouses the repulsion force to reject denser positive ions rather than the lower charged ions. Contrariwise, the divalent anions are less rejected than monovalent anions since the negative diva- lent ions experience stronger interaction with the mem- brane. These combined effects make the

5**salt rejection order to be MgCl2 > CaCl2 > NaCl > Na2SO4**

. Moreover, such a phenomenon can be explained by looking into the char- acteristics of the membrane and molecule. The larger radii of Mg2+ and Ca2+ make them held back by the membrane, while smaller Na+ ion is easier to pass through [40]. The inlet temperature (hot or cold) cause a significant difference in the desalination result. The desalination process was carried out in two beds of the adsorption sys- tem. The effect of inlet temperature is clearly explained by Elsayed et al. [43] in their study. A

3**CPO-27(Ni)** (also **known as MOF-74) and** aluminum **fumarate**

-MOF were used as the membrane modifier. Aluminum fuma- rate works by adsorbing water in the inner pores of the materials without the existence of unsaturated metal sites. Meanwhile,

> 3**the unsaturated metal sites** and **the** hydrophilic organic linker **of CPO-27(Ni**

) will provide B.L. Suwandi et al. / Desalination and Water Treatment 213 (2021) 214–228 additional binding sites for water. CPO-27(Ni), alumi- num

> 3**fumarate, and MIL-101(Cr**) exhibited **a** maximum adsorption capacity **of**

0.47 g H2O/

> 1**gads, 0.53** g H2O/**gads, and 1.47** g H2O/**gads**

, respectively [69]. By operating in the optimum condition, the CPO-27(Ni) produced freshwater 4.6 m3·1/(ton d)

> 1**at an evaporation temperature of 5°C and** a **regeneration temperature of** 110°**C**

. In comparison, alu- minum fumarate produced 6.

> 3**3 m3·(ton** d)−**1 at an** evapora- tion **temperature of** 20°**C**

and a regeneration temperature of 70°C. This study showed that

> 3**CPO-27(Ni) has a** better **performance** than aluminum fumarate **at**

high regenera- tion (desorption) temperature and low evaporation tem- perature. In comparison, aluminum fumarate

> 3**has a** better **performance at a** higher **evaporation temperature**

[43]. The effect of inlet temperature to the desalination also has been stated by Youssef et al. [70] using the same CPO-27(Ni) MOF in a 1-bed adsorption cycle. The use of CPO-21(Ni) allows maximum water output of 22.8 m3/(ton d), with the inlet temperature of 40°C for the evaporator and 5°C for the condenser. 4.2. Mechanism of desalination improvement The

> 2**presence of MOF particles in the** membrane causes the **membrane** to crack **and**

form pores around it. The more pores that form around MOF particles will provide a sig- nificant increase in surface area, thereby increasing desali- nation performance [71,72]. Without the presence of MOF, the pores

formed by the polymer are uneven – even some- times, there are areas on the membrane that have very small (or absent) pores that do not allow water permeance (Fig. 8a). By adjusting the number of embedded MOF, the pores formed will be adequate and facilitate water perme- ance (Fig. 8b). Logically speaking, increasing the number of MOFs should provide a very significant increase in the number of pores. But unfortunately, this is not the case; embedding excessive amounts of MOF can cause unwanted membrane damage (Fig. 8c). Embedding MOF beyond the capacity of the membrane will create a large membrane crack, and thus, large pores are formed, and the water (also ions) can pass freely [29,39,71–73]. Membrane pores that are too small induce a higher affinity between water molecules and pore walls; this causes high surface tension of water molecules (Fig. 9a) [74]. In this case, higher pressure is needed to enable a proper desalination process, but this is often not feasible because excessive force will cause damage to the membrane. The existence of MOF can disturb the affinity of water. MOF can help break down water molecules so that the surface tension of the water is lowered. Previous reports have shown that the use of MOF with different water-philicity provides different desalination mechanisms. Hydrophilic MOF breaks surface tension by attracting water molecules toward it (Fig. 9b), while hydrophilic MOF breaks surface tension by applying repulsive forces to water molecules (Fig. 9c). A new approach to surface-philicity design is the omniphobic membrane surface, which has excellent

such a membrane has been introduced in the MD process [75–78]. However, many trials and errors are still needed to improve omniphobic membranes; espe- cially in overcoming their weakness, which is easily dam- aged by the condensation process. Insight into the MOF performance in desalination led to the idea that adsorption capability drove the MOF per- formance. MOF adsorption capability is innate from the unsaturated metal sites that actively bind the guest mole- cules, and therefore improved the desalination efficiency. Also, MOF loading into the membrane led to porosity enlargement. The simple illustration of the desalination process using the MOF-modified and unmodified mem- brane is shown in Fig. 10. In most cases, the improvement effect (in desalination) of MOF can be further escalated by forming composites (i.e., MOF@GO). Later on, it believed that there would be more particular requirements in desali- nation membranes that can be met by the MOF-composites, such as specific or selective membrane against individual guest molecules, and thermal/magnetic/electric membranes. It can be concluded that, although the exploration of com- posite materials has just barely begun, the prospects of MOF-composite-incorporated membranes are vast and will keep on increasing [79]. The primary purpose of every sus- tainable development is the feasibility of its application in the industry. The engineers have agreed that a desalination membrane must possess specific properties to fulfill this purpose: stable (no performance declining) over a long term (a) (b) (c) Fig. 8. Illustration of pores properties of the desalination membrane. (a) Membrane with absence of MOF particles, the arrow shows the area with tiny (absent of) pores, (b) membrane with MOF particles embedded, and (c) membrane with excess MOF particles embedded, the arrows show the area with large membrane crack. B.L. Suwandi

water molecules and membrane pore walls. (a) Membrane without MOFs, (b) membrane with hydrophilic MOFs, and (c) membrane with hydrophobic MOFs. (a) (b) Fig. 10. Illustration of the desalination process. (a) Desalination by using an unmodified membrane; the pores structure is limited and (b) desalination by using MOF-modified membrane; the structural pore is enlarging thus the surface area is higher. More pores with smaller size created due to the incorporation of MOF NPs, the smaller pores give higher solute rejection. process, reusable, high efficiency, environmentally friendly, and low cost. 4.3. Prospect and challenges For the past decade,

20**RO has been the most** widely **used method for** saline water **desalination**

. However, the grow- ing development of MOF nanotechnology has proven to be able to produce high-properties membranes for desalination, such as MOF-based TFN membranes, MOF- based electrodes, MOF-based adsorbents, etc. The MOF- hybridized membranes are proven to improve desalination performance—thanks to their superior cycling stability and durable structure. The fact that MOFs apply to every possible method for desalination makes it attractive for sustainable development in this field. Nevertheless, the persistent challenges in membrane fabrication with MOFs are the achievement of superior desalination capability, high durability, minimum degrada- tion, and proper safety for humans and the environment. So far, current studies only focused on comparing the MOF embedded membrane to the plain membrane, or charac- teristic of the membrane, and improving the salt rejection and water flux in a lab-scale process. In which some of B.L. Suwandi et al. / Desalination and Water Treatment 213 (2021) 214–228 them are not feasible to implement for industrial purposes. Industrial-scale processes are required a distinctive inves- tigation and design, with their separated trials and errors. The industrialized amount of feed water, as well as salt con- centration, is needed in applicative researches as real natu- ral seawater is the targeted application. Furthermore, more research needs to be done, considering the economic and environmental sustainability of incorporating MOF in the water desalination process. 5. Conclusions Fresh and clean water scarcity has become an issue in many parts of the world. To overcome this global issue, extensive researches have been developed to establish an advanced novel technology of water desalination, with lower cost, lower energy consumption, and environmen- tally friendly. Nanomaterials such as MOFs have been used in various nanotechnology to enhance the desalina- tion process, including forward osmosis, reverse osmosis, adsorption, capacitive deionization, and membrane distil- lation. MOFs are endowed with excellent

19**properties such as high surface area, high porosity**

, high chemical stability, and feasible combination of inorganic and organic matters. The exceptionally promising efficiency improvement of desalination performance by MOF has been demonstrated throughout this review. The factors that are inducing the improvement are porosity enlargement of the membrane; additional adsorption sites provide by MOF, improved selectivity, and unique properties (e.g., magnetic). The combination of the desalination membrane with MOF offers a remarkable improvement in desalination perfor- mance, which can be a promising solution for overcoming water scarcity in the future. Still, there are some challenges in the future application of seawater desalination, that is: (1) the high energy requirements – especially for practi- cal applications, (2) the possibility of environmental dam- age during the construction of desalination equipment and piping systems, and (3) the disposal of concentrated salt water (reject-salt) which can be critical environmen- tal disturbances. Efforts in the sustainable development of the desalination process are apparent, for instance, the employment of robotic-automation technology, the use of renewable energy (e.g., solar cell) to operate the system, the development of reject-salt disposal systems (e.g., deep well injection), and the emerging of environmental regulation regarding the seawater desalination. At last, research and design on desalination systems should focus not only on developing eco-friendly and high-performance membrane materials but also on increasing cost and energy efficiency.

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