Evaluating the Performance of Fertilizer Draw Solutions by Using the Modified Cellulosic Composite Forward Osmosis Membranes

Doaa F. Ahmed[1,3,*], Heba Isawi[2], A.A. Elbayaa[3], Nagwa A. Badway[3] and Hosam Shawky[2]

[1] *Egyptian Desalination Research Center of Excellence (EDRC), Cairo, Egypt*

[2] *Desert Research Center, Water Resources and Desert Soils Division, Hydrogeochemistry Dept., Water Desalination Unit, Egyptian Desalination Research Center of Excellence (EDRC), Cairo, Egypt*

[3] *Chemistry Department, Faculty of Science, Al-Azhar University, Nasr City, Cairo, Egypt*

Abstract: This paper deals with various fertilizer influences to draw solutions to the neat CTA/CA, MA/CTA/CA, and the Al_2O_3/MA/CTA/CA nanocomposite (NC) modified membrane. Also, the applicability of the neat CTA/CA, MA/CTA/CA, and The Al_2O_3/MA/CTA/CA nanocomposite (NC) modified membrane display high water flux when it used to desalinate brine water sample collected from the brine mid-stream from Mersa Matruh area, North-Western Coast of Egypt. The salinity of the collected sample is 12760 mg/L and PH (8.5) and used as FS, and 1M from different fertilizer draw solutes (DFDS) include KCl, NH_4Cl, $(NH_4)_2SO_4$, and K_2HPO_4 used as DS. The results reveal that the flux was KCl and NH_4Cl (17.8 $L/m^2.h$) and followed by $(NH_4)_2SO_4$ (17.1 $L/m^2.h$) and K_2HPO_4 (16.6 $L/m^2.h$) using the natural saline water as FS using Al_2O_3/MA/CTA/CA NC modified membrane. The reusability test of the synthesised Al_2O_3/MA/CTA/CA NC modified membrane showed good sustainability during the 1260 min continuous test. The FO application displayed a great potential to be interested in brine wastewater desalination and enhanced water source sustainability to use in agriculture fertigation.

Keywords: Forward Osmosis Desalination, CTA/CA polymer blend, Fertilizer draws solution, brine wastewater desalination.

1. INTRODUCTION

Agricultural irrigation is exhibited as a water-intensive procedure and consumes 72% of the global freshwater extraction. Discovering alternative water resources for irrigation through reclaiming wastewater will be of great importance [1]. Water shortages have called for a significant number of researchers to pay more attention to water sustainability; Therefore, researchers have been considering highly efficient and low-energy desalination techniques [2,3]. Forward osmosis (FO, also known as direct osmosis or a natural physical phenomenon) is a promising and desirable one of the existing desalination procedures [4]. FO has been promoted as the favourite for high-water recuperation and low-cost desalination [5]. When compared with conventional pressure-energetic membrane procedures such as (Nanofiltration (NF), Ultrafiltration (UF), and Reverse osmosis (RO).

The competitive and effective FO technologies, which are the membrane material and the draw solution, are influenced by the two main keys [6]. Cellulose triacetate (CTA) is one material identified by cellulose that is abundantly available at low cost. As a result, the CTA membrane has greater salt rejection but less permeability than the CA membrane, the CTA is more hydrophobic than the CA. The CA/CTA FO composite membrane is then required to compromise the salt retention and flux of water. Consequently, one of the strongest FO membranes available for marketing is the cellulose acetate/cellulose triacetate (CA/CTA) blend membranes. Conversely, the CA/CTA blend FO membrane's use has the benefit over the (Thin-film composite) TFC complements for their favoured chlorine resistance and biodegradability [7]. The membrane surface modifications using additives can affect its performance, and the improver adding is the highest economical technique to hinder the fouling of the hydrophobic membrane. The commonly used additives are macro-molecular as polyethene glycols (PEG), polyvinylpyrrolidone (PVP), organic compounds such as glycerol, alcohols, and inorganic salts such as maleic acid, ZnCl2, acetic acid, and LiCl. The acetic acid and cis-trans isomers as pore-forming improver were blended with the CTA and CA casting mixture, correspondingly. The resultant FO membrane had a significantly more porous and open-cell sub-layer structure, leading to an improvement in the water flux and membrane possessions [8,9]. Furthermore, the pore-forming mechanism of separable pore-forming additive containing lactic acid, maleic acid, and acetic acid was examined and anticipated, which might create a complex with solvent as acetone in the casting mixture as $ZnCl_2$ and the composite may discharge

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*Address correspondence to this author at the Egyptian Desalination Research Center of Excellence (EDRC), Cairo, Egypt; Tel: (202-01014072589);
E-mail: d.fathy33@gmail.com, doaa.fatthy@sva.edu.eg

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through the phase inversion procedure [10]. Newly, significant consideration has been rewarded for the creation of nanocomposite membranes for different applications. Al_2O_3 NPs attract attention because of their stimulating physical and chemical possessions; extensively used in numerous fields such as adsorbents, composite materials, and membrane creation because they own various advantages as large pore volume, great surface area, great hydrophilicity, high porosity, and an economical price compared to CNTs, TiO_2 [11]. The Al_2O_3 NPs have an OH group on their surface, which links with the functional groups in the MA/CTA/CA polymer matrix via hydrogen or covalent bond, facilitating this combination and enhancing the constancy of the Al_2O_3 NPs into the membrane casting solution [12]. Hezma *et al*., (2019) studied the ZnO NPs were incorporated in the Cs/PVA blend films to form a nanocomposite by casting method ZnO Nps. ZnO NPs doped Cs/PVA blend shows good thermal stability, mechanical strength, and high antibacterial activity [13]. Rajeh *et al*. (2020) used The PMMA and PEMA with Co-doped ZnO NPs in electrochemical devices [14]. Hezma *et al*. (2017) studied Structural, thermal, and mechanical properties of pure blend and nanocomposites based on polyurethane (PU) and polyvinyl chloride (PVC) doped with low different content of single-walled carbon nanotubes (SWCNTs) in the field of wind turbines blades for electrical power generation [15]. Abutalib and Rajeh (2020a) prepared novel PMMA/PEO- Fe_3O_4 nanocomposite would be suitable for electromagnetic applications such as memory applications and components of hard disks [16]. (Abutalib and Rajeh, 2020c) used PEO and PMMA with MWCNTs/Li-doped TiO_2 polymer composites in electrochemical applications such as storage devices and separators in batteries [17]. Ragab and Rajeh (2020) prepared new Ag/PAM/PVA nanocomposites film's suitability for application in optical and/ or electrical devices, fabricating solid-state batteries, and other electrochemical devices[18]. Abutalib and Rajeh (2020b) studied CMC/ZnO/Co nanocomposites films in the solid-state battery application [19]. Graphene oxide sheets (GO NSs) were selected to enhance the membrane performance for forwarding osmosis (FO) application [20]. The objective of the study was to investigate the effects of pressure and pH on the neat CTA/CA, MA/CTA/CA, and Al_2O_3/MA/CTA/CA NC flat sheet. The Membranes tested using various forms of fertilizer draw solutes include; KCl, NH_4Cl, $(NH_4)_2SO_4$, and dipotassium hydrogen orthophosphate (K_2HPO_4). The neat CTA/CA, MA/CTA/CA, conducted that the FO test to assess water flux and solute reverse flux, in addition to membrane stability for FO water desalination. Additionally, the stability of the synthesised Al_2O_3/MA/CTA/CA NC modified membrane related to neat CTA/CA, and MA/CTA/CA membranes will test using the FO lab-scale for water desalination. The permeate water flux experiment prepared using DI waters as FS and 1 M NaCl as DS with AL-FS at 25°C while the membrane performances test completed using 0.1M NaCl as FS, and $1MNH_4Cl$ as DS with AL-FS at 25°C. and to evaluate the overall feasibility of FO the neat CTA/CA, MA/CTA/CA, and Al_2O_3/MA/CTA/CA NC flat sheet in the brine wastewater with as a makeup water source of agriculture. Bywater sample collected from the brine mid-stream from Mersa Matruh area, North-Western Coast of Egypt. The salinity of the collected sample is 12760 mg/L and PH (8.5). The salinity of the selected water sample is 12760 mg/L, were used as a feed solution, and 1M of various kinds of draw solutes include; KCl, NH_4Cl, $(NH4)_2SO_4$, and K_2HPO_4 used as draw solutions with the operative layer site of the membrane faces the feed solution reservoirs.

2. EXPERIMENTAL

2.1. Materials

The membrane substrate was prepared from a mixture of cellulose triacetate (CTA) with an averaged acetyl content of 43–44 wt%, and cellulose acetate (CA) with 39.7 wt% acetyl content (Mn=30,000) was purchased from Acros Organics (USA). A mixture of 1,4-dioxane and acetone with purity 99.5 and 99.9.5%, respectively, were used as a solvent. Maleic acid (MA) with purity 99% has become from Sigma-Aldrich. Aluminium oxide (Al_2O_3) nanoparticles (NPs) with nano-size 10 nm were used as an additive and provided by Sigma Aldrich. Sodium chloride (NaCl) salt, potassium chloride (KCl), dipotassium hydrogen orthophosphate (K_2HPO_4), ammonium chloride (NH_4Cl), and ammonium sulfate $(NH_4)_2SO_4$ were obtained from ADWIC, Nasr Pharma, Egypt and used as draw solutions. The distilled water (DI) was provided from an ultra-pure water model (pure lab Option-K method, UK), using a resistivity of >15 mΩ-cm.

2.2. Preparation of Modified Nanocomposite flat-Sheet Al_2O_3/MA/CTA/CA-Based Membranes

The CA and CTA powders were dried over 70-80 ° C to a constant weight using an oven. The neat CTA/CA membrane has been created through the phase inversion technique. The tidy CTA/CA membrane was cast with an equivalent polymer content of 14 Wt from homogeneous polymer solutions. CTA/CA (1:2) per cent powder and 86 Wt. As in our previous publication, the percentage of a 1,4-dioxane/acetone (3:1) mixture as a solvent [12]. After that, the casting polymer solutions were cast onto a dry and clean glass plate using a casting knife of 200 μm and speed of $0.01 m\cdot s^{-1}$ using a TQC Automatic film applicator. After solidification and peel off, the membranes were immersed in DI water that was changed every four h for 24 h. After preparation and annealing at $85^{o}C$ for 5 min to eliminate the excess solvent. The optimised composition of the polymer solution was fixed at (14:60:26) (1CTA:2CTA): dioxane: acetone, 5.25 wt. % MA, and 0.2 wt. % Al_2O_3 NPs cast for further application on the non-woven support layer (SL).

2.3. Membrane Performance Assessment for Forwarding Osmosis (FO)

The FO performance of the resulting neat CTA/CA, MA/CTA/CA, and Al_2O_3/MA/CTA/CA NC changed membranes in salt rejection. Water flux expressions were calculated in the FO model with 1L DI H_2O as FS, and 1M NaCl solution was used as DS and/or 1 M NaCl solution as feed solution (FS) and 1M ammonium chloride (NH_4Cl) solution as draw solution (DS). The test cell involves two rectangular plastic symmetric half cells with the membrane in between, Figure **1**. The half-cell measurements are 12.7 cm in length, 10 cm in width, and 8.3 cm in height. The membrane was mounted in a module with a membrane area of 42 cm^2 (CF042A-FO), allowing the discharge solution and the feed solution to flow counter at present on each membrane outline at the same velocity of 1.8 L/min as in the case of the discharge solution and the feed solution. Figure **1**. FO performance in expressions of water flux and salt retention of the resultant prepared membranes was estimated. Both FS and DSs were circulated using gear pumps. The concurrent FS and DS flow were adopted to decrease the strain on the postponed membrane. The flow ratio was determined to be 1.6 L/min after an optimisation test to reduce the external concentration polarisation. A circulator maintained both FS and DSs at $25^{\circ}C \pm 0.5^{\circ}C$. Automatic thermometers with the correctness of $0.1^{\circ}C$ were utilised to identify the solution temperature at the entry and way out of the module. FO performance mostly was evaluated below the FS model (the FS faced the opaque eclectic layer and the DS faced the support layer) over 1 h, via calculating reverse solute flux (RSF, J_s) and permeate water flux (J_v). Considered data were reserved after an initial stabilisation period of about 10 min. Water flux was considered via the weight alteration of the DS owing to the water permeation as follows:

To estimate the FO permeates water flux (Jw, $L/m^2.h$), the volume of water (ΔV) transferred from the FS to the DS was measured and applied in the next equation:

$$J_w = [V_w / (Am \times t)] \quad (1)$$

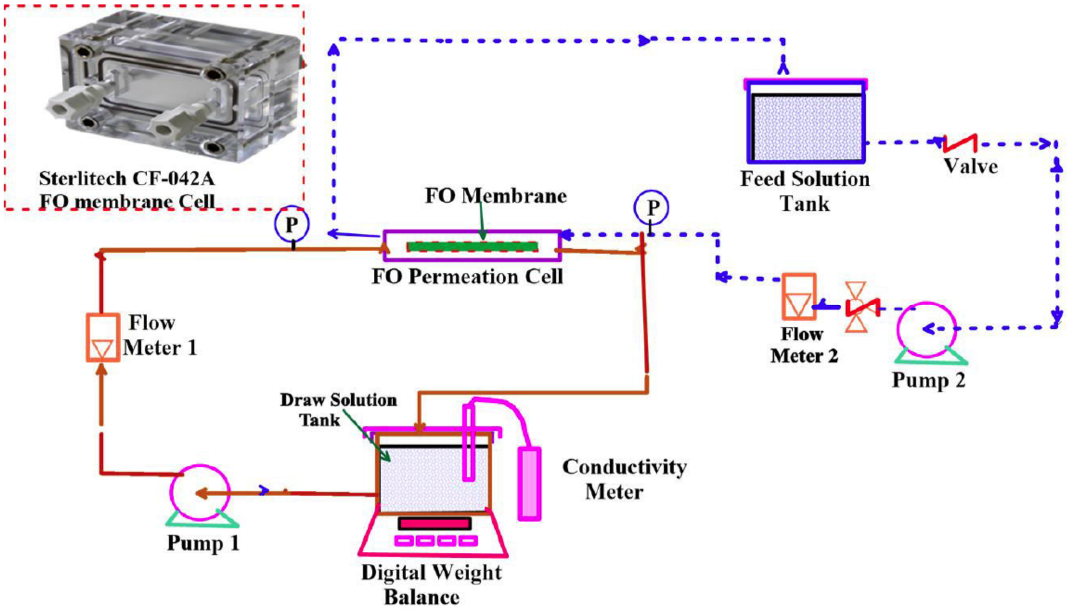

Figure 1: Forward Osmosis experimental setup for FO membrane test and Sterlitech CF-042A FO membrane Cell.

Where V_w is the permeate water volume (L) passed over the membrane surface area A (m^2), t is the purification time (h). The inverse salt flux (Js, g /m^2. h) was determined via;

$$J_S = (V_t x C_t – V_0 x C_0) /(\Delta t \times A_m) \quad (2)$$

Where V_o and V_t are the starting, and final volumes of the feed solution and C_o and C_t are the starting and ending salt concentrations of the FS. The trials were finalised at 25°C. The salt rejection (R, %) was considered as below;

$$R= 1 – [C_{NaCl} X V_{DS} /C_{FS0} X V_{FS0}] X100 \quad (3)$$

Where $C_{NaCl, ds}$ is the salt concentration in the DS after a period of time, which was estimated via standard curve method using a conductivity meter (DDS-307a), and V_{ds} is the volume of the draw solution. C_{fs0} and V_{fs0} are the initial concentration and volume of the feed solution, respectively.

3. RESULTS AND DISCUSSION

3.1. Performance Evaluation of FO Membranes

3.1.1. The Effect of Different Concentration of NaCl as a Draw Solution

The FO performance of the fabricated neat CTA/CA, MA/CTA/CA, and Al_2O_3/MA/CTA/CA NC modified membranes were evaluated in the form of water flux, and RSF experiments were performed using DI as FSs and different concentration of various from 0.5 and 3M of NaCl as DSs, flow rate 1.6 L/min, operation temperature 25°C and the membrane operational area of 42 cm^2, Figure **2**.

Figure **2** shows the water flux and the RSF of neat CTA/CA, MA/CTA/CA, and Al_2O_3/MA/CTA/CA NC modified membranes as a function of the DSS concentration (NaCl). The water flux and RSF increased gradually with increasing content from 0.5 to 3M. The water flux increases to 30,33, and 35 L/m^2.h, while reverse solute flux little increase to1.04, 6.02, and 11.09 for the cellulose acetate membrane, MA/CTA/CA membrane, and AL_2O_3/MA/CTA/CA membrane, respectively (at constant operating conditions). The higher the water flux of the cellulose acetate membrane, MA/CTA/CA, and Al2O3/MA/CTA/CA membrane indicating the higher the membrane porosity. As can be seen, the water Flux increases for each membrane with increasing DS concentration due to increased osmotic driving force. Furthermore, higher water flux was consistently observed in the modified nanocomposite membranes compared with the neat membrane [20]. At higher DS concentrations, the accumulation of salt ions increased ICP and reduced water flux. Besides, the two effects of ICP and membrane fouling decreased osmotic water flux and increased mass transfer resistance as the feed water becomes more concentrated during FO permeation time [12]. This is caused by the higher severity of ICP in the FO mode, resulting from the dilution of draw

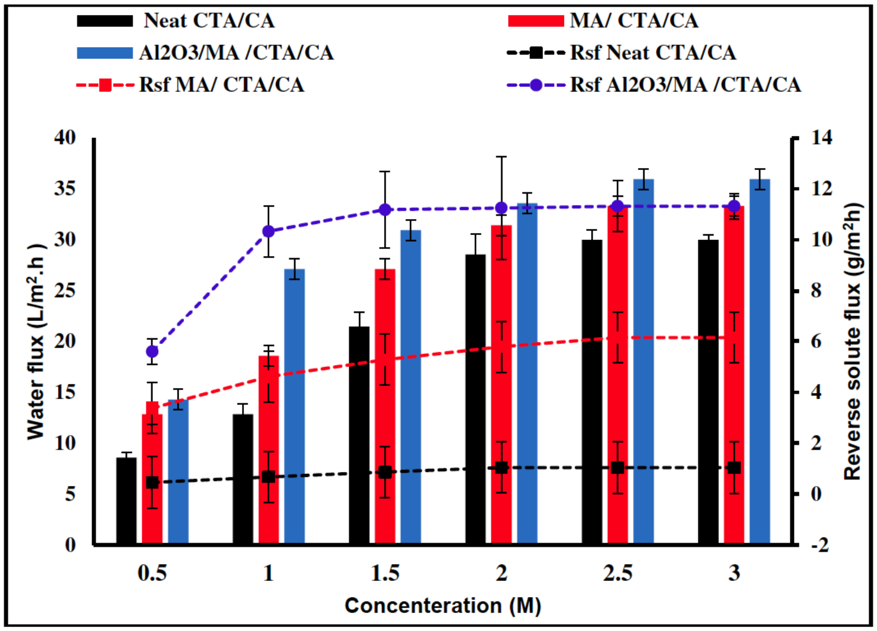

Figure 2: The effect of different NaCl concentration as a DSs on water flux, and reverse solute flux of neat CTA/CA, MA/CTA/CA, and Al_2O_3/MA/CTA/CA NC modified membranes DI water as FSs on AL- FS mode.

solution within the porous substrate at a higher draw solution concentration [21]. Because of the greater concentration gradient across the membrane, higher reverse salt flux was also observed at higher DS concentrations. As shown in Figure **2**, both MA/CTA/CA and Al_2O_3/MA/CTA/CA NC membrane experienced slightly higher reverse salt flux than that of the neat membrane at different DS. We have obtained a higher water flux and acceptable solute reverse flux. This result demonstrates that Al_2O_3 nanoparticles would be a potential nanomaterial to fabricate high-performance FO membranes [12]. RSF increased as DS concentration increased due to the salt concentricity gradient growth over the membrane sides that provide a higher driving force for the migration of NaCl ions from the DS side to the FS side. A similar observation was made in previous studies [22]. In other words, the DS concentricity gradient is the driving strength for the simultaneous passage of H_2O and salt through the membrane in the opposite direction. We have obtained a higher water flux along with an increase in the RSF, presenting typical trade-off phenomena. A higher RSF always occurred when there was higher water penetration via the membrane [23].

3.1.2. Effect of Applied Pressure on the Performances of Forwarding Osmosis Membranes

The water flux and reverse solute flux experiments were performed using DI as feed solution and 1M NaCl as draw solution, flow rate 1.6 l/min operation temperature 25°C, the effective membrane area of 42 cm^2, applying a hydraulic pressure on the feed side from 0 to 2 bar, and with AL-FS membrane orientation Figure **3**.

Figure **3** shows water flux and reverse solute flux (RSF) of neat CTA/CA, MA/CTA/CA, and Al_2O_3/MA/CTA/CA NC membrane at different operating pressures. Water flux and reverse solute flux increased gradually with increasing applied pressure from 0 to 2 bar. The water flux increases to 26.6, 33.33, and 38.09 $L/m^2.h$, while reverse solute flux little change to 1.48, 4.9, and 11.2 $g/m^2.h$ for the neat CTA/CA, MA/CTA/CA, and Al_2O_3/MA/CTA/CA NC modified membrane, respectively (at a constant operating condition). Improvement of water permeation flux was observed when applying a hydraulic pressure on the feed side, leading to significant permeation flux enhancement in AFO (Assisted forward osmosis) application [24]. Firstly, the water flux increases due to the increased driving force causing in feed ion concentrations in water permeate flux and hydrophilicity of the modified membrane [24,25]. A positive impact of the applied hydraulic pressure on the water permeation flux was observed [26]. The decrease in RSF may be attributed to physical changes in the membrane active layer as a function of the hydraulic pressure. Also, increasing pressure at the membrane surface may have compressed the interface between the thin active layer and a support layer of the CTA/CA membranes, thus reducing RSF of the membrane, while minimally affecting water flux the lower RSF is attributed to the elevated salinity of the feed in the current experiments

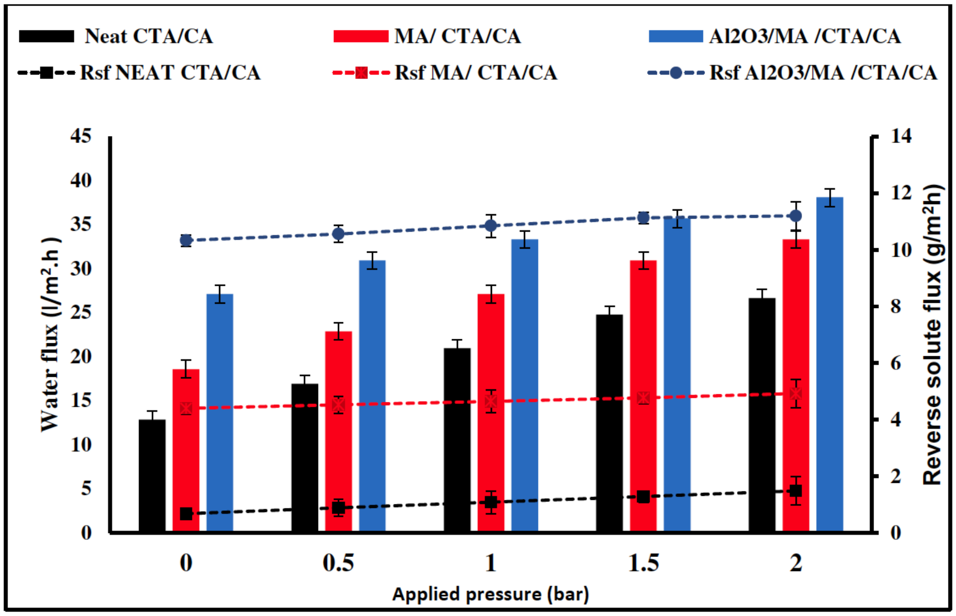

Figure 3: The effect of applied pressure on the on-water flux and reverse solute flux of (a) neat CTA/CA, MA/CTA/CA, and Al_2O_3/MA/CTA/CA NC modified membranes DI water as FSs on AL- FS mode.

coupled with increased concentration polarisation expected with high water flux [27].

3.1.3. Effect of Feed pH on the Performance

The FO performance of the fabricated neat CTA/CA, MA/CTA/CA, and Al_2O_3/MA/CTA/CA NC modified membranes were evaluated in the form of water flux, and RSF experiments were performed using DI with different pH as FSs and 1M NaCl as DSs, flow rate 1.6 L/min, operation temperature 25°C and the membrane operational area of 42 cm^2, Figure **4**. To investigate the effects of feed solution pH on the membrane performance, 0.1 mol/L NaOH and 0.1 mol/L HCl were added to the feed solutions to adjust pH Figure **4** shows the water flux and the RSF of neat CTA/CA, MA/CTA/CA, and Al_2O_3/MA/CTA/CA NC modified membranes as a function of the different pH as FSs. The water flux and RSF increased slightly with increasing pH from 3 to 12. The water flux increase to 13.5,19.2, and 27.8 L/m^2.h, while reverse solute flux little increase to 3.6, 4.4, and10.75 for the cellulose acetate membrane, MA/CTA/CA membrane, and AL_2O_3/MA/CTA/CA membrane, respectively (at constant operating conditions). The water flux increased by 12.2, 13.5, and 14.5% in the neat CTA/CA, MA/CTA/CA, and Al_2O_3 FO membrane, respectively, was observed with the increase of the feed solution pH from 3 to 12. This behaviour may be attributed to conformational changes of the cross-linked membrane polymer structure and changes in the membrane hydrophobicity as a function of the solution pH [28]. It is also noted that the FO membrane surface becomes more hydrophilic through the dissociation of carboxyl functional groups (COO^-) of the active layer as the solution pH increased. A more hydrophilic membrane could favour water transport. This hypothesis is consistent with the correlation between hydrophobicity of FO membrane and water flux observed by McCutcheon and Elimelech [29]. The reverse solute fluxes with different neat CTA/CA, MA/CTA/CA, and Al_2O_3 FO membrane presented in Figure **1**. The results revealed that the reverse solute flux was affected by the feed solution pH. The reverse salt flux increased with increasing feed solution pH (Figure **4**). This implies that better selectivity and efficiency are expected with a basic feed solution rather than that with an acidic one. This increase in the reverse salt flux is mostly driven by the increase in water flux. The water flux increases significantly at higher pH whilst the salt flux is suppressed by the more negatively charged FO membrane. Therefore, the electrostatic exclusion didn't contribute to the reverse solute flux, resulting in a constant reverse solute flux over the entire pH range. In comparison with the previous study [28]. Zhang *et al*., 2017 studied the TFC FO membrane had a lower reverse salt flux than the CTA FO membrane, which was in agreement with the information provided by the manufacturer [29].

3.1.4. Influence of Different Fertilizer Draw Solution Properties on the Performance of the FO Process

In this section, the influences of the different fertilizer DS properties were investigated on the performance of the FO desalination process using neat CTA/CA, MA/CTA/CA, and Al_2O_3/MA/CTA/CA NC modified membranes. Different types of fertilizer draw solutes include KCl, NH_4Cl as monovalent ions,

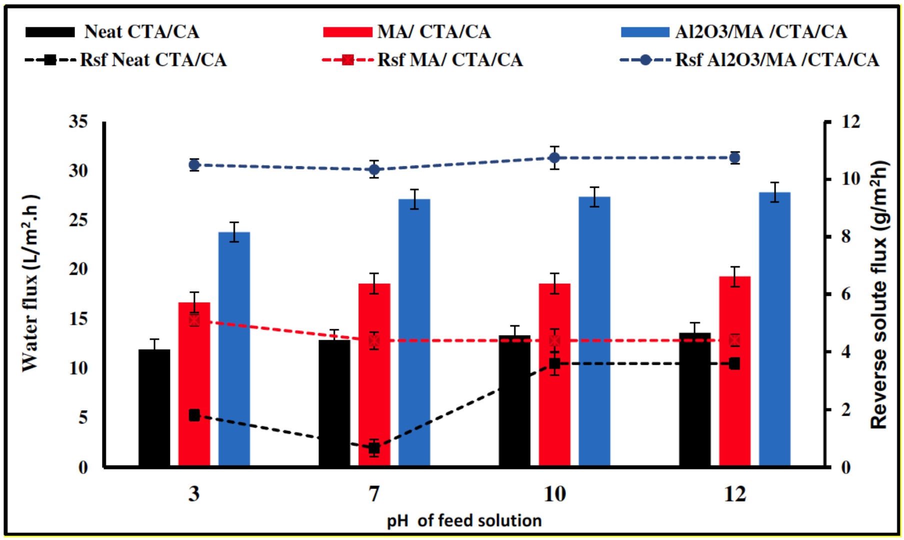

Figure 4: The effect of different pH feed solution on water flux and reverse solute flux of neat CTA/CA, MA/CTA/CA, and Al2O3/MA/CTA/CA NC modified membranes DI water as FSs on AL- FS mode.

$(NH_4)_2SO_4$ as divalent cations and dipotassium hydrogen orthophosphate (K_2HPO_4) as divalent anion using DI as FD and 1M from each different fertilizer DSs, flow rate 1.6 l/min, concentration operation temperature 25°C and the effective membrane area of 42 cm^2, Figure **5a**. These fertilizers draw solutions were selected depending on their charges, shape, and molecular weight, Table 1. Kind of DS plays an essential function in the performances of the FO besides its osmotic pressure. The selected membranes were applied in the FO performances of four various fertilizers DSs as examples for the desalination of brine H_2O for irrigation; the chemical features for the chosen draw fertilizer were obtainable in Table **1**. Figure **5a** shows pure water flux J_w of each fertilizer DS as a function using neat CTA/CA, CTA/CA/MA, and Al_2O_3/MA/CTA/CA NC modified membranes. The membranes possess water flux values in the following order: KCl=NH_4Cl> $(NH_4)_2SO_4$> K_2HPO_4 using neat CTA/CA, CTA/CA/MA, and Al_2O_3/MA/CTA/CA NC modified membranes. The results showed that the Al_2O_3/MA/CTA/CA NC modified membrane has a higher water flux compared to neat CTA/CA, CTA/CA/MA membranes using the selected various DSS. The greater water flux using KCl and NH_4Cl than the $(NH_4)_2SO_4$ and K_2HPO_4 can be attributed to the higher diffusion coefficient of monovalent electrolytes as compared to bivalent electrolytes and trivalent electrolyte [30]. DS can extract H_2O from the FS tank when the osmotic pressure of the DS reaches stability with the osmotic pressure of the FS [30]. The performance of fertilizer DSs in terms of RSF varied widely depending on the type of fertilizers, also is shown in Figure **5a**. The K_2HPO_4 and $(NH_4)_2SO_4$ showed the lowest RSF compared to the other selected fertilizers KCl and NH_4Cl. The results indicate that the fertilizer DSs containing monovalent elements have higher RSF than divalent elements [$(NH_4)_2SO_4$ and K_2HPO_4]. The divalent fertilizer draw solutions $(NH_4)_2SO_4$ and K_2HPO_4 have ionic species with a hydrated diameter of PO_4 and SO_4 relatively much greater than the hydrated diameter of the monovalent fertilizer species (NH_4, Cl, and K) and then might be unique of the aims for lower RSF, Table 2. Furthermore, RSF might be particularly important, especially when phosphorus and nitrogen, including DSS, are utilised as these composites are identified to cause eutrophication in the getting water environment. Consequently, it is vital to calculate the performance of fertilizer DSs concerning RSF. All of the membranes verified were oriented so that the AL faced the FS, though a greater water flux is attained when the DS is on the effective side; since ICP is less severe, operating with the effective side facing the feed is recommended when the feed water has a high fouling potential [30]. The membrane flat sheet model demonstrated the best performance, particularly water flux, particularly at high DSs osmotic pressures.

To evaluate each fertilizer DS's performance of each fertilizer DSs, FO process performance experiments were taken using 1 M concentration DS and 0.1M NaCl as FS, and their water flux measured Figure **5b**. The neat CTA/CA membrane has water flux values in the following order: KCl (12.6 L/m^2.h)> NH_4Cl=$(NH_4)_2SO_4$ (12.3 L/m^2.h)> K_2HPO_4 (11.9 L/m^2.h) using neat CTA/CA. While in the case of the MA/CTA/CA membrane, the NH_4Cl (18.5 L/m^2.h) has the highest water flux, followed by $(NH_4)_2SO_4$ (17.1 L/m^2.h) and K_2HPO_4 (16.6 L/m^2.h). The Al_2O_3/MA/CTA/CA NC modified membranes showed the highest water flux using KCl and NH_4Cl (26 L/m^2.h) and followed by $(NH_4)_2SO_4$ (24 L/m^2.h) and K_2HPO_4 (21 L/m^2.h). Figure **5b** Showed the salt rejection for each fertilizer DS as a function using neat CTA/CA, MA/CTA/CA, and Al_2O_3/MA/CTA/CA NC modified membranes. K_2HPO_4 has the highest salt rejection compared to NH_4Cl,$(NH_4)_2SO_4$, and KCl. These results might be due to the significance of fertilizer DS molecular weight that plays an essential role in the FO procedure. Smaller fertilizers draw molecules, KCl and NH_4Cl exhibit identical separation styles than $(NH_4)_2SO_4$ and K_2HPO_4 where $(NH_4)_2SO_4$ and K_2HPO_4 were rejected higher than the NH_4Cl and KCl. The molecular size and the chemical origin of natural inorganic matter are valuable tools for evaluating their effects on the performances of FO desalination organisations. The FO separation efficiency depends on the charges on the membrane surfaces and the natural inorganic matter that plays an extensive role in the rejection process. The $(NH_4)_2SO_4$ and K_2HPO_4 have the same trend for FO rejection due to the greater molecular weight of their chemical structure where the divalent fertilizer draws solutions $(NH_4)_2SO_4$ and K_2HPO_4) have higher retention than the monovalent ones. The result found that the rejection of $(NH_4)_2SO_4$ and K_2HPO_4 was higher than the solutes and membranes. The outcomes indicate that K_2HPO_4 fertilizers are the important group of the greatest concentrated soluble fertilizer materials containing both k and P elements essential fertilizer components wanted by the plants and can be either alone as fertilizer or is favourable to be used as mixed fertilizer with several other fertilizers. Although their water fluxes

Table 1: Chemical Characteristics for the Selected Draw Solutions

Name of fertilizers	Chemical Formula	MW	π at 1.0 M (atm)	π at 1M calculated π = n. C.R.T (atm)
Ammonium chloride	NH_4Cl	53.5	43.5	49.26
Potassium chloride	KCl	74.6	44.0	49.26
Ammonium sulphate	$(NH_4)_2SO_4$	132.1	46.1	73.89
Dipotassium hydrogen orthophosphate	K_2HPO_4	174	36.5	73.89

The bulk osmotic pressure (π) value, according to Phuntsho *et al*., 2012.

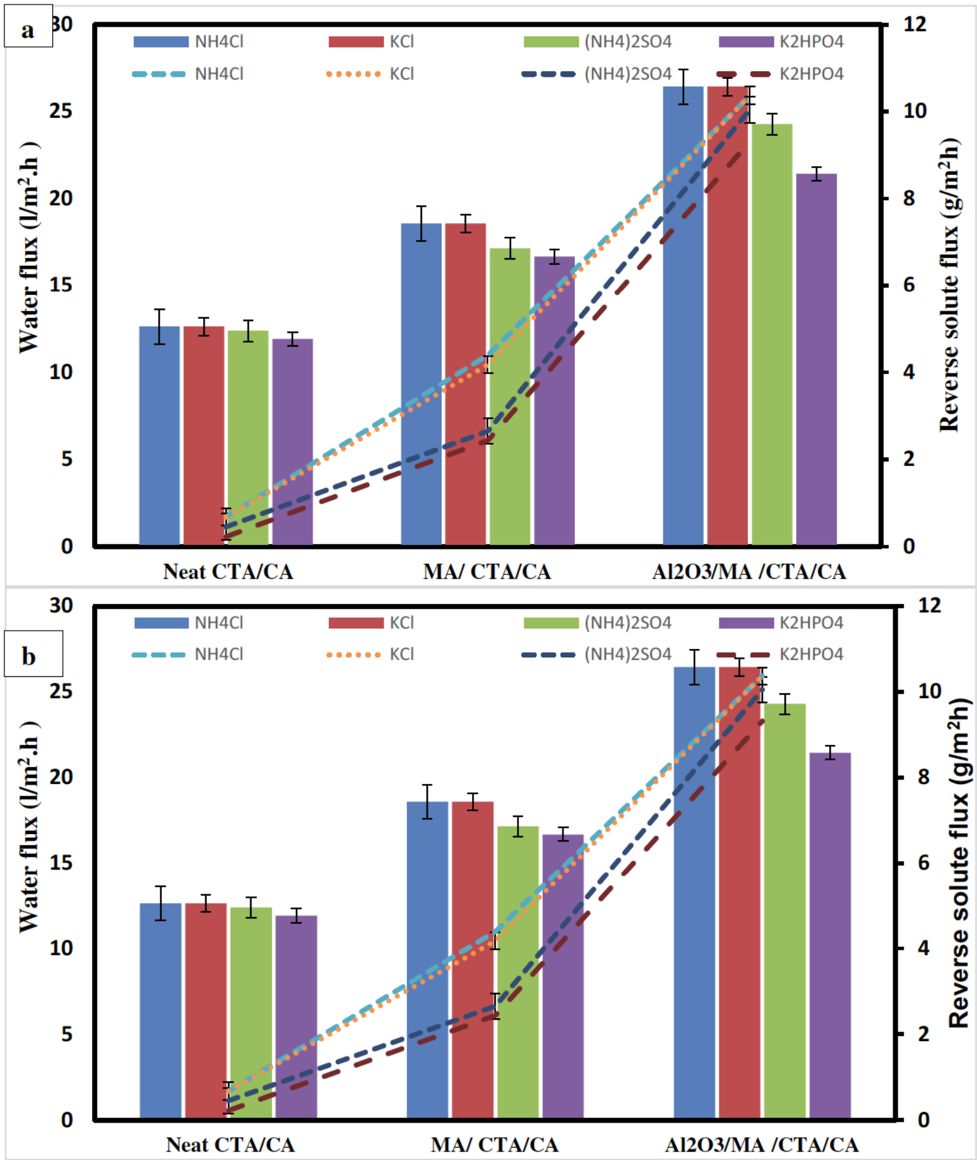

Figure 5: (**a**) Influence of the different type of fertilizer DS on the performances of FO process in terms of water flux and reverse solute flux using DI as FS and 1M from each selected draw solutions; (**b**) Water flux and salt rejection using 0.1M NaCl as FS and 1M from each selected draw solutions as KCl, NH_4Cl, $(NH_4)_2SO_4$ and K_2HPO_4, (AL-FS), temperature 25 °C, flow rate 1.6 l/min, using neat CTA/CA, MA/CTA/CA and Al_2O_3/MA/CTA/CA NC modified membranes. 1.6 L/min flow rate, with cells vertically oriented and, evaporation time 2 min; water bath temperature 12 °C.

are lower than the other draw solutions, their low RSF makes them a promising candidate for FO desalination for fertigation using Al_2O_3/MA/CTA/CA NC modified membrane.

3.2. Application of FO Synthetic Membranes in a Natural Brine Wastewater Sample

The productions of fresh water from saline water are one of the extremely substantial challenges facing Egypt at present, as Egypt does face not only a water scarcity problem but also an inevitable energy crisis. The neat CTA/CA, MA/CTA/CA, Al_2O_3/MA/CTA/CA NC modified membranes were selected for desalination of a natural saline water sample collected from the Mersa Matruh area, North-Western Coast of Egypt. The Mersa Matruh area has a dry climate where the freshwater is insufficient to meet the probable increases in water demands. This study selected four types of fertilizer as the DS using the neat CTA/CA, MA/CTA/CA, Al_2O_3/MA/CTA/CA NC modified FO membranes for the desalination of a natural saline water sample with TDS (12760 mg/L) and PH (8.5). Mersa Matruh region deliberated to be one of the greatest promising localities for land reclamation in Egypt. Mersa Matruh is located in the southern portion of the Mediterranean coastal semi-arid area and has been exposed to an intensive estimation by governmental authorities for the establishment of the new communities, tourist societies, and land reclamation developments. The sample properties, prior FO process, are shown in Table 2. To study the performances and the desalination capacity of the synthesised membranes, so the necessary laboratory tests have been achieved, and membranes tested. The FS contains a natural brine wastewater sample with the total dissolved solids 12760 mg/L and 1M from different types of FDSs include KCl, NH_4Cl, $(NH_4)_2SO_4$, and K_2HPO_4 as DSs, flow rate 1.6 L/min, at $25^{\circ}C$ and the membrane operation area as 42 cm^2 with FS-AL mode. The FS reservoirs are placed on weighing balances until the stability of the salt rejection and water flux has reached. Previously, The neat CTA/CA, MA/CTA/CA, Al_2O_3/MA/CTA/CA modified membranes showed high performances. Included the water flux and salt rejection using the synthesised solution from 0.1M NaCl and DI as FS, and 1M from different types of FDSs include KCl, NH_4Cl, $(NH_4)_2SO_4$, and K_2HPO_4 used as a DS, Figure **5a**, and **b**. An evaluation study was evaluated in Figure **6**, where the water flux and salt rejection of the selected natural saline water sample collected from the Mersa Matruh area as FS. where the water flux and salt rejection of the selected natural saline water sample collected from the Mersa Matruh area as FS. The amount of measured water flux of the neat CTA/CA membrane showed that KCl and NH_4Cl have the highest water flux (9.1 L/m^2.h) followed by $(NH_4)_2SO_4$ (8.8 L/m2.h) and K_2HPO_4 (7.1 L/m2.h). In the case of the MA/CTA/CA membrane, the water flux showed the highest value as in order KCl and NH_4Cl (13.09 L/m^2.h)> $(NH_4)_2SO_4$ (12.3 L/m^2.h)> K_2HPO_4 (11.9 L/m^2.h) followed the same trend in the case of the Al_2O_3/MA/CTA/CA NC modified membranes that display the highest water flux as in order KCl and NH_4Cl (17.8 L/m^2.h)> $(NH_4)_2SO_4$ (17.1 L/m^2.h)> K_2HPO_4 (16.6 L/m^2.h) Figure **6**.

Flux in an FO experiment can be influenced by the concentration of DS and FSs, as well as by the overall osmotic pressure variance (Δπ) among the DS and the FS and fouling of membrane [32,33]. The primary driving force for flux in FO can consider as a variance in osmotic pressure between the FS and DS. Therefore, the osmotic pressure variance between the 0.1 M NaCl and the DS was lower than the saline water sample as an FS and DS, as represented in Figure **5**. Owing to the combined high TDS altitudes and the organics, the activity or “effective concentration” of constituents in the natural brine water sample was higher than that for the salt copy. Moreover, the natural saline water sample contains a large number of ions with high TDS shown in Table 2. The membrane fouling includes both organic fouling and inorganic fouling (i.e., scaling) anticipated being a key reason for the severe flux decrease shown in Figure **6**. In this work, the FO rejections (R %) of the FS were evaluated by taking the DS sample at the finale of every test and analysing it for Na+ ions Figure **6** and Table **3**. All desalted permeated water samples collected for chemical analyses after one hour. Figure **5**, showed the salt retention for each fertilizer DS as a function using The neat CTA/CA, MA/CTA/CA, Al_2O_3/MA/CTA/CA NC modified membranes. K_2HPO_4 has the highest salt rejection compared to NH_4Cl, $(NH_4)_2SO_4$, and KCl for all membranes. These results might be owing to the impact of fertilizer DS molecular weight that plays an essential role in the FO process. Smaller fertilizers draw molecules, KCl and NH_4Cl exhibit identical separation styles than $(NH_4)_2SO_4$ and K_2HPO_4 where $(NH_4)_2SO_4$ and K_2HPO_4 were rejected higher than the KCl and NH_4Cl. The full chemical analyses of the feed and the product (permeate) water samples were revealed in Table 2. When using NH_4Cl as DS, it is obvious that the concentricity of the cations and anions decreases after the desalination procedure in Table 2. The rejection per cent (R%) has measured using the brine wastewater sample as FS, Table 3. The Al_2O_3/MA/CTA/CA modified membranes show higher water flux compared to The neat CTA/CA, MA/CTA/CA,

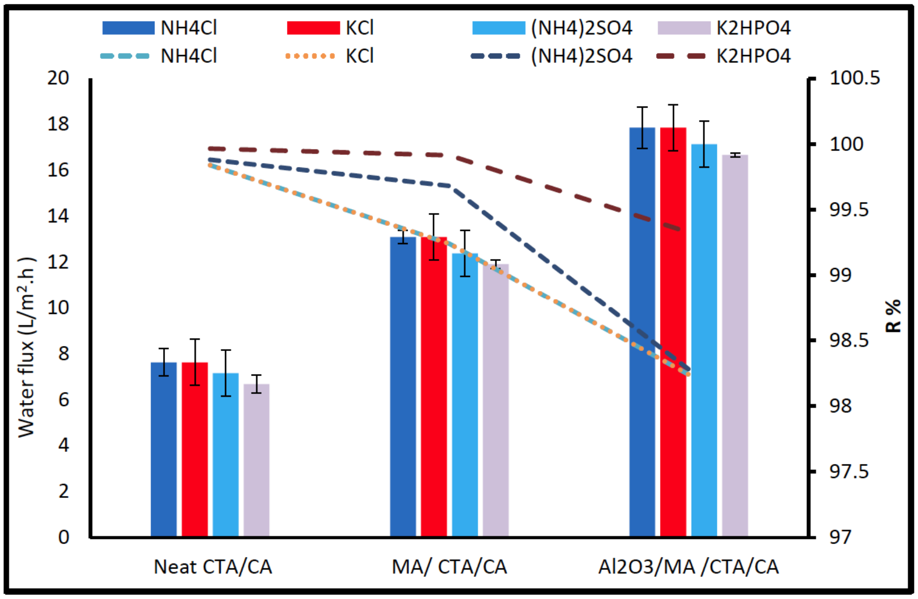

Figure 6: Influence of the different types of DSs on the performances of FO process in terms of water flux and salt rejection using a natural saline water sample collected from Mersa Matruh desalination plant North-Western Coast of Egypt, using different draw solutions as KCl, NH_4Cl, $(NH_4)_2SO_4$ and K_2HPO_4 at 25 °C, flow rate1.6 L/min, FO mode: with cells vertically oriented, and (AL-FS), using The neat CTA/CA, MA/CTA/CA, Al2O3/MA/CTA/CA.

Table 2: Characteristics of a Natural Saline Water Sample Selected from Mersa Matruh Area, before and after FO Desalination Using Neat CTA/CA, AA/CTA/CA, and GO/AA/CTA/CA NC Modified Membranes and NH_4Cl as a Draw Solution

Ion	Concentrations (mg/L) of brine water before FO system.	Concentrations (mg/L) of the brine wastewater effluent were collected from the Mersa Matruh area, after the FO system.		
		CTA/CA	MA/CTA/CA	Al2O3/MA/CTA/CA
TDS	12760	12880	12932	13045
Ca^{+2}	296	297	298	299
Mg^{+2}	708	709	710	711
Na^{+}	3366	3465	3500	3600
K^{+}	107	108	108	109
CO_3^{-2}	120	120	120	120
HCO_3^{-}	134	134	134	134
SO_4^{-2}	1049	1050	1052	1053
Cl^{-}	7046	7066	7076	7085

membranes. The highest water flux of Al2O3/MA/CTA/CA NC modified membranes achieved due to the OH, COOH, and epoxy functionalised modified membrane. This high performance is anticipated because the H_2O molecules could be simply drawn into the membrane surface and bulk by enhancing the hydrophilicity of the membrane.

3.2. Effect of Time on Activity of Synthesised FO Membranes

Fouling raises power consumption and effects in a great operating cost. Inorganic compounds can be adsorbed or deposited onto the effective layer of the membrane, causing membrane fouling, which in turn decreases the flux [33]. Figure **7** shows the effect of time on water flux of neat CTA/CA, MA/CTA/CA, and Al_2O_3/MA/CTA/CA NC modified membranes with the (AL-FS) mode using brine wastewater as the FS and 1 M NH_4Cl as the DS. The outcomes indicate that the flux decreased gradually with time through FO membrane processes via osmotic pressure as the driving power for water flux via the membrane. It is found that the water fluxes for the Al_2O_3/MA/CTA/CA NC modified membrane were higher than the water

Table 3: The Reversions Flux and Salt Rejection of Saline Water Sample Selected from Mersa Matruh Area, before and after FO Desalination Using The Neat CTA/CA, MA/CTA/CA, Al_2O_3/MA/CTA/CA Modified Membranes Using NH_4Cl as a DSS

Ions	Saline water sample before (mg/L)	The revers ions Flux (g/m^2.h)			R%		
		Neat CTA/CA	MA/CTA/CA	AL2O3/MA/CTA/CA	Neat CTA/CA	MA/CTA/CA	AL2O3/MA/CTA/CA
Ca^{+2}	296	6	5	6	99.82	98.3	97.9
Mg^{+2}	708	2.8	5	6	99.05	99.2	99.1
Na^{+}	3366	6	20	49	99.82	99.4	98.5
K^{+}	107	4	2	2	98.64	98.1	98.1
CO_3^{-2}	120	3	6	5	99.57	99.15	99.29
HCO_3^{-}	134	10	20	16	99.85	99.71	99.77
SO_4^{-2}	1049	1	3	2	99.06	97.19	98.13
Cl^{-}	7046	22	24	26	97.90	97.7	97.5

fluxes of neat CTA/CA, MA/CTA/CA membranes. The flux kept declining as a result of the dilution of draw solution and concentricity of FS. Consequently, the flux decay in the fouling tests is produced not only via fouling of the membrane but also via the decrease in osmotic driving strength [32]. The FO membrane fouling reversibility was owing to the lower dense organic fouling coating created in the FO style because of the hydraulic pressure shortage or the back diffusion of ions from the DS to the membrane surface on the feed sideway as described.

The enhancement of fouling resistance of Al_2O_3/MA/CTA/CA NC modified membrane is mainly due to the developed membrane surface hydrophilicity as well as the higher surface negative charges afterwards incorporation of Al_2O_3 NPs. The Al_2O_3/MA/CTA/CA NC modified membrane has the highest fouling resistance with a final water flux value of 16.04 L/m^2.h about 80 % of the initial value compared to neat CTA/CA, MA/CTA/CA membrane which has 5.66 and 10.65 L/m^2.h about 45 and 74% of their initial values, respectively. This indicated that the Al_2O_3 NPs embedded membrane exhibit an enhancement in CTA/CA membrane hydrophilicity and might hinder the solute deposition and adsorption on the membrane matrix. After introducing the MA as a pore-forming agent and incorporation of hydrophilic Al_2O_3 NPs to the CTA/CA membrane, thus the membrane became more hydrophilic, so the membrane fouling was delayed owing to the enhancement of the membrane surfaces with the hydrophilic MA besides the polymer matrix of the FO membranes used in this study are made of CTA/CA, which is known to have relatively small fouling potential. When DI water was used as the FS, water flux in FO style is higher than 0.1M NaCl, and the saline water sample used as the FS under the same flow rate (1.6 L/min) and DS (1M) concentration Table 3. These outcomes propose that fouling plays a more pronounced role in the flux pattern when a real saline water sample is used. Both external concentration polarisation (ECP) and internal concentration polarisation (ICP) also vary with the changing DS/FS concentrations. High FS concentricity also contributed to the increase in ECP that directly caused the high reduction of the resultant water flux [34]. This result illustrated that the concentration polarisation (CP) affects the DS and FS concentrations at the membrane AL and causes a lower actual flux in the FO, as revealed in Table 4. ICP and ECP, along with the dilution and concentration of DS/FS, participated in the reduced available osmotic pressure across the membrane's AL, which resulted in a lower flux outcome compared with the high-theoretical flux potential. The FO membrane always gives a flux driven by the concentrations present at the membrane surfaces and not by the actual contents of DS and FS. As a result of DS/FS concentration variance at the prepared membrane interface, the resultant lower FO flux based on the $\Delta\pi$ effective across the prepared membrane interface instead of the flux based on the $\Delta\pi$ bulk, i.e., the variance in osmotic pressure among the DS and FS themselves.

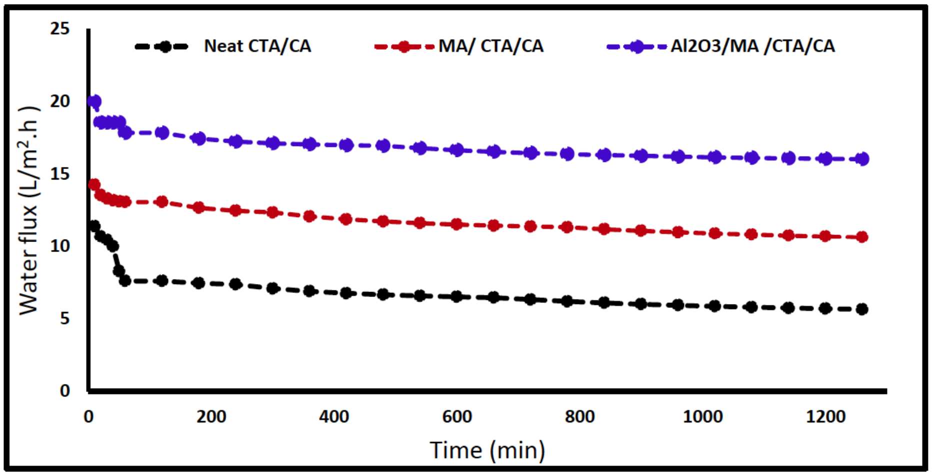

Figure 7: Permeate flux versus filtration time for neat CTA/CA, MA/CTA/CA, and Al_2O_3/MA/CTA/CA NC modified membranes. 1.6 L/min flow rate, with cells vertically oriented, and (AL-FS), evaporation time 2 min; water bath temperature 12°C.

Table 4: The Water Flux of The Neat CTA/CA, MA/CTA/CA, Al2O3/MA/CTA/CA NC Modified Membranes Using Different Fertilizer as DSs and DI water, 0.1 M NaCl, and the brine wastewater Sample Selected from the Mersa Matruh Area, Respectively as an FSs, after FO Desalination

Membrane types		**Neat CTA/CA**	**MA/CTA/CA**	**Al_2O_3/MA/CTA/CA**
Draw solution types		**Water Flux (L/m^2.h)**		
NH_4Cl	Feed (DI)	12.6	18.57	26.42
	Feed (0.1M NaCl)	12.6	18.57	26.42
	Feed (Brine)	9.1	13.09	17.85
KCl	Feed (DI)	12.6	18.57	26.42
	Feed (0.1M NaCl)	12.6	18.57	26.42
	Feed (Brine)	9.1	13.09	17.85
$(NH_4)_2SO_4$	Feed (DI)	12.6	17.14	24.28
	Feed (0.1M NaCl)	12.4	17.14	24.28
	Feed (Brine)	8.8	12.3	17.14
K_2HPO_4	Feed (DI)	11.9	16.66	21.41
	Feed (0.1M NaCl)	11.9	16.66	21.41
	Feed (Brine)	7.1	11.9	16.6

It revealed that the Al_2O_3/MA/CTA/CA modified membranes show higher results compared to neat CTA/CA, and MA /CTA/CA membranes for saline water desalination using the FO technique. Figure **7** shows Fertilizers were proposed as DS to extract water from natural saline water samples for direct irrigation. The diluted fertilizers can be applied directly without any further treatment as a source of nutrients for irrigation. Utilising the FDFO for the desalination of the saline water sample is an efficient technique that reduces the amount of salt from the saline water without applying any hydraulic pressure. Thus, this is an ecologically friendly technique that saves energy and keeps the environs and lower desalination cost. FO is a promising alternative sustainable technology promoting saline water desalination The FDFO technique is in not requiring a recovery step to re-concentrate the DS so, the alternative using the diluted DS as a supplement for irrigation H_2O by fertigation.

4. CONCLUSION

The current study improved CTA/CA blends flat sheet membranes via incorporating various amounts of MA as a pore-forming agent The results showed that the Al_2O_3/MA/CTA/CA NC modified membrane has a higher water flux compared to neat CTA/CA, CTA/CA/MA membranes using the selected various

DSs. The Al_2O_3/MA/CTA/CA NC modified membrane presented higher water flux 27.1 L/m^2.h and low reverse solute flux 99.1% when compared to neat CTA/CA and MA/CTA/CA membranes using 1M NH_4Cl as DS and 0.1M NaCl as FS under the AL-DS mode. This outcome is above 2 times greater than the FO water flux of the neat CTA/CA and MA/CTA/CA membranes. Using different types of fertilizer DSs include KCl, NH_4Cl as monovalent ions, $(NH_4)_2SO_4$ as divalent cations and K_2HPO_4 as divalent anion using DI as FD and 1M from each different fertilizer DSs. The Al_2O_3/MA/CTA/CA NC modified membrane showed the highest water flux using KCl and NH_4Cl (26 L/m^2.h) and followed by $(NH_4)_2SO_4$ (24 L/m^2.h) and K_2HPO_4 (21 L/m^2.h) as well as the K_2HPO_4 and $(NH_4)_2SO_4$ showed the lowest RSF compared to the other selected fertilizers KCl and NH_4Cl. The results indicate that the fertilizer DSs containing monovalent elements have higher RSF than divalent elements ($(NH_4)_2SO_4$ and K_2HPO_4). Applicability of the Al_2O_3/MA/CTA/CA NC modified membrane displays the highest water flux using the selected the brine mid-stream from Mersa Matruh area, North-Western Coast of Egypt with salinity 12760 mg/L and PH (8.5) and used as FS, and 1M from different fertilizer draw solutes (DFDS) include KCl, NH4Cl, (NH4)2SO4, and K2HPO4 used as DS. The results reveal that the flux was KCl and NH4Cl (17.8 L/m2.h) and followed by (NH4)2SO4 (17.1 L/m2.h) and K2HPO4 (16.6 L/m2.h). The reusability test displays that the synthesised Al_2O_3/MA/CTA/CA NC modified membrane showed sustaining performance stability in the 1300-min continuous desalination experiments. The synthesised Al_2O_3/MA/CTA/CA NC modified membrane will have a wide application in FO procedure for brine wastewater desalination as a result of their enhanced structural and separation possessions.

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