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Selection of potential fertilizer draw solution for fertilizer drawn forward osmosis application in Egypt

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ABSTRACT

Fertilizer drawn forward osmosis desalination success is greatly affected by the choice of a suitable draw solution. This study focused only on nitrogenous-based fertilizers being by far the most dominant class of fertilizers used in Egypt. Four nitrogenous Egyptian fertilizers have been closely evaluated with respect to their availability, economics and performance. The three factors played a major role in the fertilizer selection. Ammonium sulpahte was selected to be the most suitable fertilizer draw solution exhibiting high osmotic pressure, being non-expensive, non-hygroscopic, resistant to valorization, highly soluble in water and containing sulphur, which is needed by the plant.

Keywords: Draw solution; Egypt; Fertigation; Fertilizer drawn forward osmosis; Forward osmosis

1. Introduction

The choice of a proper draw solution (DS) is vital in forward osmosis (FO) desalination process [1,2]. Exploration of different kinds of draw solutes and choosing the optimum DS attracted many researchers in the past decades. A review of different DSs and their recovery techniques is shown in Table 1.

A DS could be any aqueous solution with high osmotic pressure. It should provide sufficient force to cause passage of water across the membrane, and therefore, it is an essential part of the FO process [28]. As the osmotic pressure of the DS is the driving force in the FO, it is crucial to select an appropriate concentrated solution for any application [29]. The osmotic pressure relies on concentration, number of species generated, the molecular weight of the solute and the temperature [4]. Osmotic pressure is independent on the type of species generated in the solution (colligative property). The less the molecular weight of the DS and the higher its water solubility, the more the osmotic pressure generated and the higher the flux [11].

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2. Draw solution selection criteria

An efficient draw solute must have the following distinctive properties [11]:

- It must exhibit a high osmotic driving force.
- It has to be soluble in water.
- It preferably has a small molecular weight.
- It must be non-toxic.
- It must be chemically matched with the membrane.
- The DS solute should be easily and inexpensively recovered (if not needed in the product water).

A flow diagram that displays the DS selection criteria is shown in Fig. 1.

2.1. Fertilizers as draw solutes

The choice of fertilizer DS for fertilizer drawn forward osmosis (FDFO) application will be based on a number of factors that are fertilizer availability, economics and performance.

Table 1
Summary of the draw solutions tested in FO investigations and their recovery techniques [3–5]

Year(s)	Draw solute/solution	Recovery method	Reference
1964	Ammonia and carbon dioxide	Heating	[6]
1976	Glucose–fructose	None	[7]
1989	Fructose	None	[8]
1992	Glucose	Low pressure RO	[9]
1997	$MgCl_2$	None	[10]
2005–2007	NH_3 and CO_2 (NH_4HCO_3) or NH_4OH–NH_4HCO_3	Moderate heating (~60°C)	[11,12]
2007	Dendrimers	Adjusting pH or UF	[13]
2007	Albumin	Denatured and solidified by heating	[13]
2008	Salt, ethanol	Pervaporation-based separations	[14]
2010	2-methylimidazole based solutes	Membrane distillation (MD)	[15]
2010	Magnetic nanoparticles	Recycled by external magnetic field	[16,17]
2011	Stimuli-responsive polymer hydrogels	Deswelling of the polymer hydrogels	[18,19]
2011	Hydrophilic nanoparticles	UF	[20]
2011	Fertilizers	None	[21]
2011	Fatty acid-polyethylene glycol	Thermal method	[22]
2012	Sucrose	NF	[23]
2012	Polyelectrolytes	UF	[1]
2012	Thermo-sensitive solute (derivatives of Acyl-TAEA)	Not studied	[24]
2012	Urea, ethylene glycol, and glucose	Not studied	[25]
2012	Organic salts	RO	[26]
2012	Hexavalent phosphazene salts	Not studied	[27]
2014	Hydro acid complexes	Recycled	[3]

2.1.1. Fertilizer availability

To have a sustainable FDFO process, the selected fertilizer should be readily available in the local market. Preferably, the fertilizer would be locally produced to avoid problems and delays related to importing from abroad. Being a central aspect of the system, fertilizer scarcity would significantly affect process efficiency.

2.1.2. Fertilizer economics

Current fertilizer prices are related to high demand due to an increasing worldwide need for more food and a more diverse diet. Fertilizer is a world market commodity subject to global market forces, volatility and risks. Yet, as the fertilizer is a key component of the FDFO scheme, for FDFO to be cost effective, the chosen fertilizer should not be expensive or costly.

2.1.3. Fertilizer performance

The selected fertilizer should have suitable physiochemical properties to serve as a DS in FDFO process, such as solubility, pH compatibility with selected FO membrane, molecular weight, osmotic pressure, water extraction capability and final nutrient content in product water [29]. In addition, the DS should not chemically react with the feed solution (FS) to create unwanted species impeding the osmotic process or the final intended utilization of the produced water (irrigation in case of FDFO).

2.2. Fertilizers in Egypt

Although there are many types of chemical fertilizers used in agricultural industry in many parts of the world, only those fertilizers commonly used in Egypt were considered for assessment as DS for FDFO. In addition, chemical composition of commercially available blended fertilizers remains proprietary, and thus, they were excluded in this work.

For Egypt, fertilizer existed a long time ago. Fertilizers are divided into two groups: organic and inorganic fertilizers [31], where the latter are used intensively in Egypt compared with the former. Inorganic fertilizers include three main categories that are nitrogen, phosphate and potassium fertilizers. Fig. 2 illustrates the percentages of fertilizers consumed in Egypt by type. It is claimed that more than 8.5 million tons (86% of total fertilizers) of nitrogenous, 11.3 million tons (11%) of phosphorus and 29 million tons (3%) of potassium fertilizers are used in Egypt [32]. Thus, nitrogen fertilizers come to be the most consumed type of fertilizer in Egypt, and this group includes urea, ammonium nitrate, ammonium sulphate and calcium nitrate. Local consumption of nitrogen fertilizers increased by 14.3% in 2008 compared with 2004. Presently, the annual use for nitrogen fertilizers is almost 9 million ton [33].

Fertilizer-use in Egypt boomed during the last three decades. For instance, in 2002 the total fertilizer consumption exceeded 1.3 million tons [34]. Fig. 3 illustrates production, import, exports and consumption of different fertilizers types in Egypt. There are 14 major Egyptian

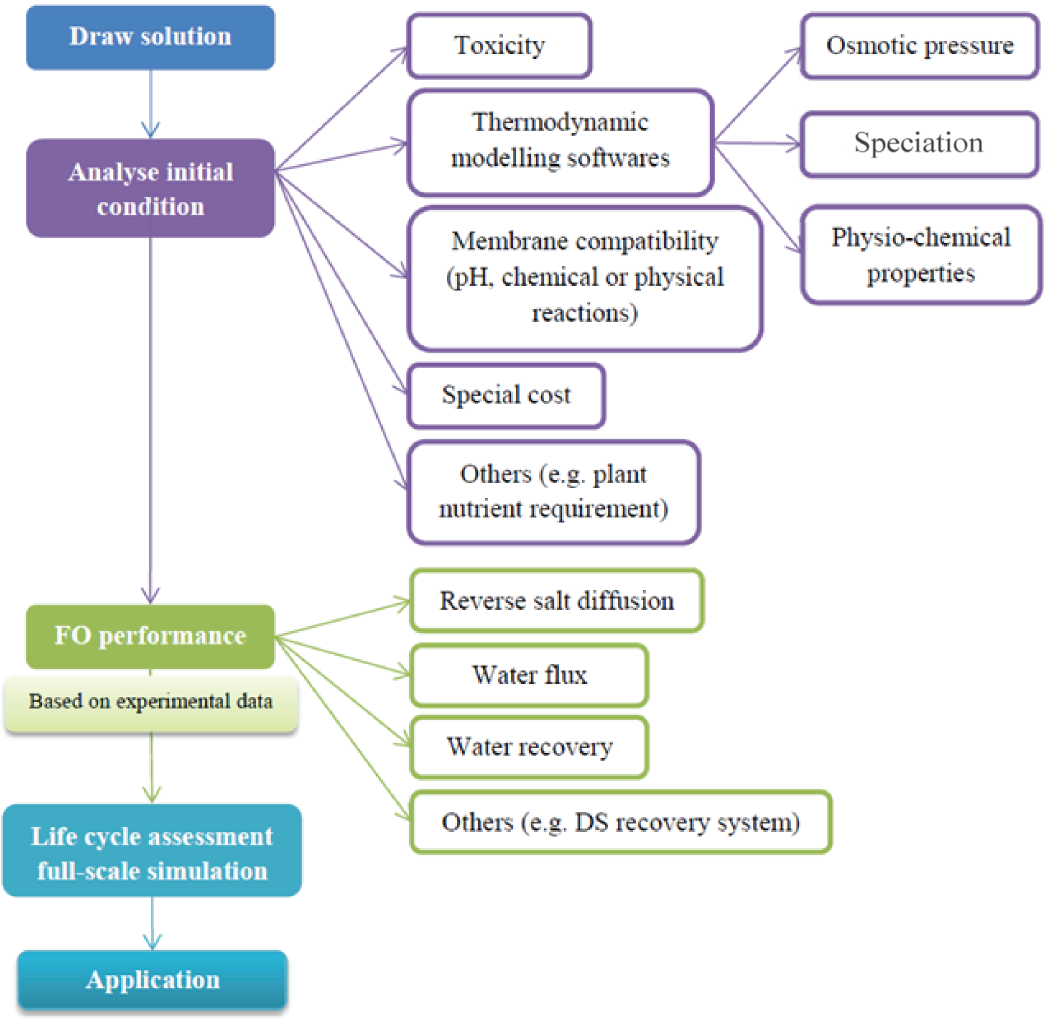

Fig. 1. Flow diagram for selecting a suitable DS in FO process [30].

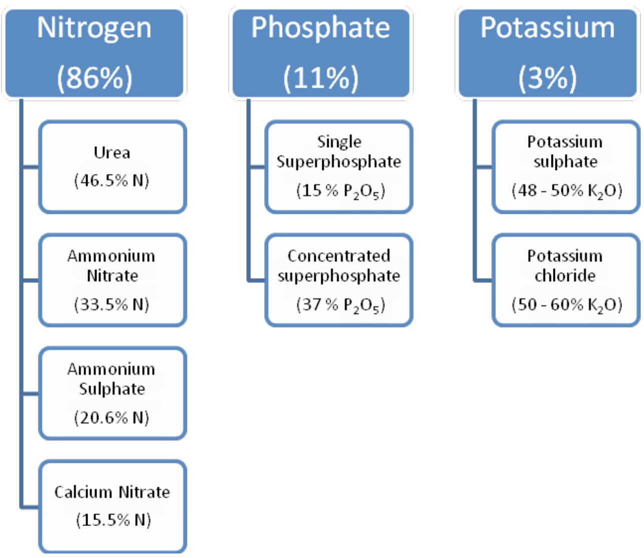

Fig. 2. Main types of fertilizers Egypt. Amounts presented are consumption percentages [34].

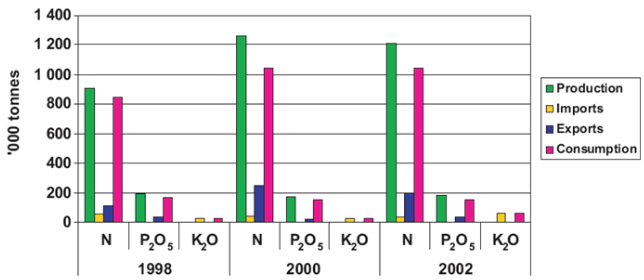

Fig. 3. Production, imports, exports and consumption of fertilizers in Egypt [34].

fertilizer-producing companies such as Semadco, Abu Qir Co., Abu-Zaabal Fertilizer and Chemical Company and others [35].

As nitrogenous fertilizers are by far the most commonly produced and consumed fertilizers in Egypt, this investigation will focus only on them (Figs. 2 and 3).

2.3. Fertilizer screening according to availability

The four selected fertilizers are available in the market (Fig. 2). Yet, nitrate containing fertilizers (ammonium nitrate and calcium nitrate) were not easy to obtain as they are categorized as explosive material.

2.4. Fertilizer screening according to economics

Average local market prices of both highly pure and less pure fertilizers have been collected from different suppliers. Prices of highly pure (99% purity) chemical fertilizers were used for comparison. As each fertilizer contains a different amount of nitrogen content, comparison is carried out on a kilogram of nitrogen basis, as per Table 2. Urea contains the highest nitrogen content (46%) followed by ammonium nitrate, ammonium sulphate and calcium nitrate. While urea contains the largest percentage of nitrogen, it is considered more expensive (in terms of kg N) than calcium nitrate and ammonium sulphate.

The prices of the four selected fertilizers are illustrated in Fig. 4. Ammonium nitrate is the most expensive fertilizer costing 462 LE/kg, followed by calcium nitrate, urea and ammonium sulphate. The order changes if the basis for comparison is kilogram of N as follows: ammonium nitrate followed by urea, calcium nitrate and ammonium sulphate.

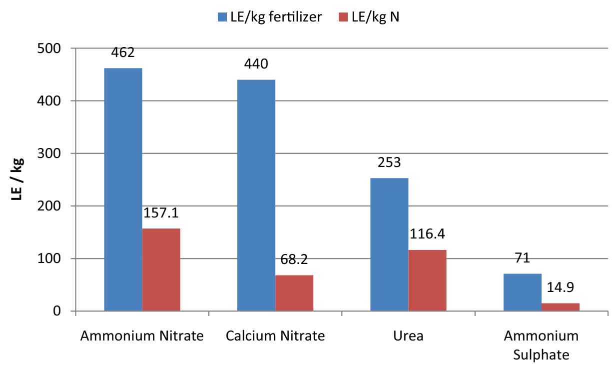

Fig. 4. Price comparison of four selected fertilizers [36].

2.5. Fertilizer screening according to performance

A performance screening of nitrogen-based fertilizer for the DS is conducted to determine basic properties (Table 2). OLI Stream Analyzer software 9.1, a software that employs thermodynamic modelling from published experimental data to forecast properties of solutions at different concentrations, was used to determine DS solubility, pH, speciation and osmotic pressure.

2.5.1. Osmotic pressure

The osmotic pressure relies on the number of species produced rather than the species' nature [38]. Fig. 5 shows the osmotic pressure of the four selected fertilizers DS at variable concentrations. Calcium Nitrate produces the largest osmotic pressure of 600 atm at its maximum solubility. This is because $Ca(NO_3)_2$ when dissolved generates the largest number of species in comparison with other fertilizers.

If a comparison is made at the same molar concentration (say at 2.0 M) from Table 3, the next maximum osmotic pressure observed is for ammonium sulphate (92.1 atm). The least osmotic pressure witnessed is for urea (46.1 atm at 2.0 M). Yet,

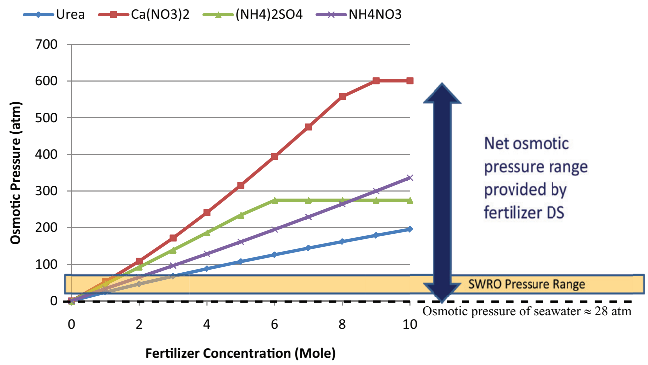

Fig. 5. Osmotic pressure of different nitrogenous fertilizers DS at 25°C analyzed using OLI Stream Analyzer 9.1.

Table 2
Fertilizer price comparison

Fertilizer	Less pure fertilizer price (LE/kg)	Highly pure fertilizer price (LE/kg)	% Nitrogen	Pure fertilizer price (LE/kg N)
Urea	2.8	253	46%	116.4
Ammonium nitrate	3	462	34%	157.1
Ammonium sulphate	1.9	71	21%	14.9
Calcium nitrate	3	440	15.5%	68.2

Table 3
List of most popular nitrogenous fertilizers in Egypt. Solubility and osmotic pressure data obtained from OLI Stream Analyzer software 9.1 [37]

Name of fertilizer	Chemical formula	Molecular weight	pH at 2 M	π at 2 M (atm)	Max. solubility
Urea	$CO(NH_2)_2$	60.05	7.00	46.1	19.65 M
Ammonium nitrate	NH_4NO_3	80.04	4.87	64.9	Highly soluble
Ammonium sulphate	$(NH_4)_2SO_4$	132.1	5.46	92.1	5.7 M
Calcium nitrate	$Ca(NO_3)_2$	164.1	4.68	108.5	7.9 M

as urea is readily soluble in water, it possesses osmotic pressure more than 200 atm at concentrations more than 10 M (Fig. 5). Figs. 5 and 7–9 provide the type and concentration of each species present as well as the expected osmotic pressure at different concentrations of the four selected fertilizers. Analysis was done by the help of OLI Stream Analyzer 9.1 software.

It is worth noting that seawater reverse osmosis (SWRO) pressure range is between 60 and 100 atm and that the osmotic pressure of seawater is estimated to be around 28 atm [39–41]. Comparing these values to the osmotic pressures of the four fertilizers under study, it is clearly inferred that the four fertilizers possess osmotic pressure much more than that of seawater and SWRO (Fig. 5).

For ammonium sulphate, three dominant aqueous species exist that are ammonium ion, sulphate ion and ammonium sulphate ion. Ammonia and bisulphate ion are not considered from the dominant species (Fig. 6). Osmotic pressure of ammonium sulphate seems to increase as concentration increases up to 5.5 molar concentration due to its maximum solubility.

Urea has only one dominant aqueous species (Fig. 7). The osmotic pressure lineally increases as urea concentration increases. Osmotic pressure reaches up to 150 atm at a 7 molar concentration.

For ammonium nitrate, two dominant aqueous species exist that are ammonium nitrate and ammonium ion. Ammonia and nitrate ion are not considered from the dominant species (Fig. 8). Osmotic pressure of ammonium nitrate seems to increase proportionally as concentration increases reaching 230 atm at 7 molar concentration.

Calcium nitrate has three dominant aqueous species that are nitrate ion, calcium ion and calcium mono-nitrate ion (Fig. 9). Osmotic pressure of calcium nitrate seems to increase proportionally as concentration increases reaching 475 atm at 7 molar concentration.

Any draw solute should exhibit higher osmotic pressure than that of the FS. For example, seawater has an osmotic pressure of 28 atm. Therefore, if seawater is the FS, the DS must exhibit an osmotic pressure a lot more than 28 atm.

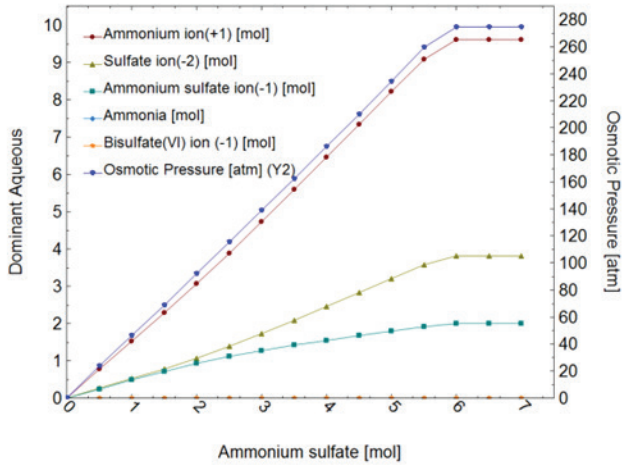

Fig. 6. Species generated and osmotic pressure of ammonium sulphate. Analysis carried out using OLI stream Analyzer 9.1 at 25°C temperature and 1 atm pressure [37].

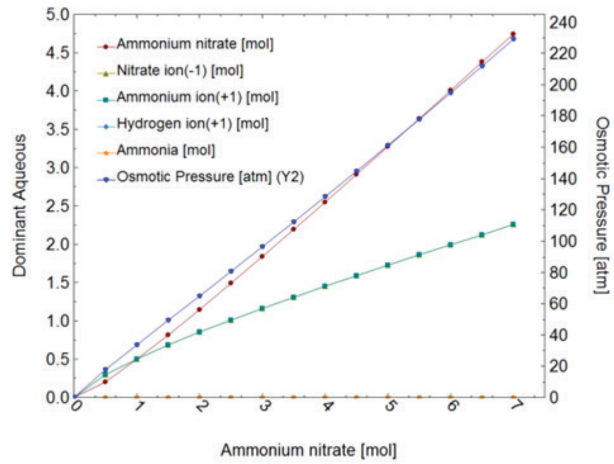

Fig. 8. Species formed and osmotic pressure of ammonium nitrate. Analysis carried out using OLI stream Analyzer 9.1 at 25°C temperature and 1 atm pressure [37].

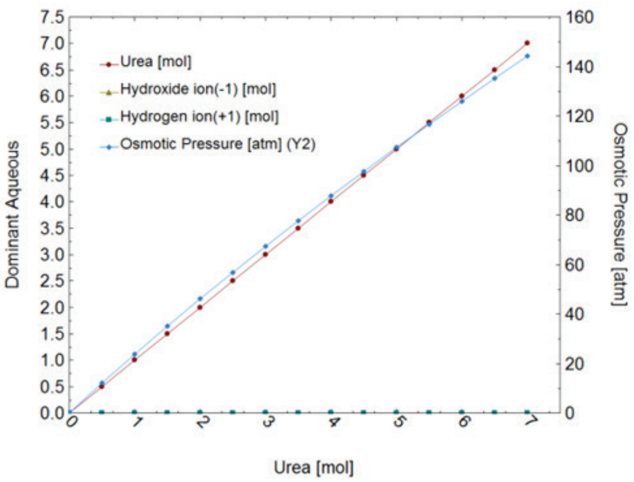

Fig. 7. Species formed and osmotic pressure of urea. Analysis carried out using OLI stream Analyzer 9.1 at 25°C temperature and 1 atm pressure [37].

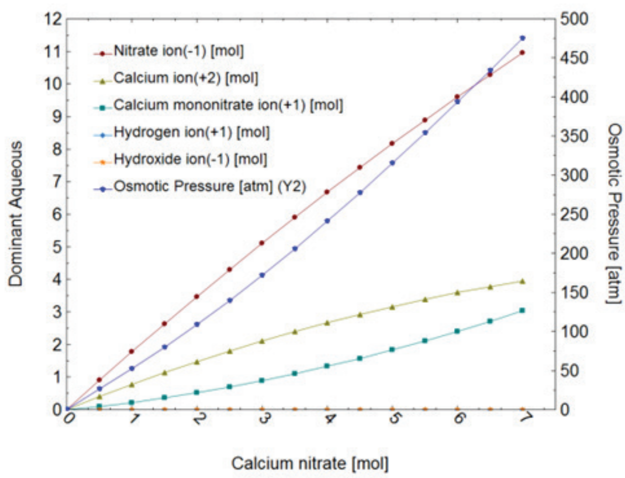

Fig. 9. Species formed and osmotic pressure of calcium nitrate. Analysis carried out using OLI Stream Analyzer 9.1 at 25°C temperature and 1 atm pressure [37].

Such conclusion signifies that all the investigated fertilizers produce osmotic pressure that is much more than that of seawater or brackish water, indicating their suitability for use as an osmotic DS.

2.5.2. Water extraction capacity

Water extraction capacity of the draw solute plays a major role in any FO process. DS can extract water from the FS until the osmotic pressure of the DS reaches equilibrium with the osmotic pressure of the FS [42]. When different draw solutes are used, a number of species are formed in solution, and the osmotic pressure of the DS depends on their osmotic coefficient. According to Phuntsho et al. [42], the total volume of water (V) a kilogram of draw solute can extract from an FS can be estimated using the following relationship:

$$V = \frac{1000}{M_w}\left[\frac{1}{C_{D,E}} - \frac{1}{C_{D,Max}}\right] \quad (1)$$

where M_w is molecular weight of draw solute used (mol/g) – Table 3; $C_{D,E}$ is the molar concentration of the DS that generates equal bulk osmotic pressure (osmotic equilibrium condition) with the osmotic pressure of a FS (mol); and $C_{D,Max}$ is maximum solubility of the draw solute (mol) – Table 3.

Osmotic pressure of six different TDS FS are considered for comparative reasons (1, 2, 5, 10, 20 and 35 g/l NaCl). Using OLI Stream Analyzer 9.1, the osmotic pressures of these FS were estimated to be 0.8, 1.59, 3.91, 7.76, 15.52 and 28 atm, respectively.

For example, to calculate volume of water extracted using urea DS and a 5 g/l NaCl FS, $C_{D,E}$ is first estimated. The 5 g/l NaCl FS has osmotic pressure equal to 3.91 atm, and the equivalent concentration of urea at this osmotic pressure ($C_{D,E}$) is equal to 0.1607 M (Fig. 7). OLI Stream Analyzer software 9.1 was utilized in these calculations. Substituting the relevant values in 1, the volume of water extracted will equal 103 L/kg.

As per Fig. 10, the water extraction capacity of the DS declines severely upon gradual increase in feed total dissolved solids. It can also be concluded that the four fertilizers almost show similar water extraction capacities. Yet, NH_4NO_3 exhibits slight more water extraction especially at low TDS feeds. For example, at a feed TDS equal to 1 g/l NaCl, while NH_4NO_3 extracts 700 l/kg of pure water, $Ca(NO_3)_2$ extracts only 488 l/kg. As FS concentration increases from 1 to 35 g/l NaCl, the difference in extraction capacities of the four fertilizers significantly decreases.

2.5.3. Expected final nutrient concentration in product water

Regardless of which initial DS concentration is used, the FO process will continue to take place until the osmotic pressure of the diluted DS is in equilibrium with the FS. Thus, the molar concentrations of each fertilizer DS can be determined according to the osmotic pressure of the FS. The feed waters of six different TDS (1, 2, 5, 10, 20 and 35 g/l NaCl) are considered to assess the expected nutrient content in the final product water after desalination.

The nutrient content is assessed in terms of nitrogen content and is presented in Fig. 11. For example, urea's final concentration at 5 g/l NaCl as FS (osmotic pressure equal to 3.91 atm) is expected to be 0.1607 M. This concentration of urea contains $\left(0.1607\frac{\text{mol}}{l}\times 28\frac{g}{\text{mol}}\right)$ g/l of N, or 4.5 g/l of N.

It is obvious from Fig. 11 that the final nutrient concentrations in FDFO rely on the type of fertilizer used and the TDS of the FS. Feed TDS and final nutrient concentration of product water are directly proportional. The lowest N concentration was observed for $Ca(NO_3)_2$, with 349 mg/L with feed TDS of 1 g/L; however, this increases to 0.72, 1.87, 3.89, 8.2 and 14.8 g/L of N with 2, 5, 10, 20 and 35 g/l feed TDS, respectively. Urea will result in highest N content in the final product water for all feed concentrations. These results indicate that when high N containing fertilizers such as urea are used as DS, the N content in the product water will be considerably higher than in the other fertilizers containing low nitrogen [43]. Another reason for high N concentration with urea is that it generates one of the lowest osmotic pressures amongst all the fertilizers at equimolar concentration, in spite of its high solubility (Fig. 11 and Table 3).

2.5.4. Dilution requirement

If the final product water from the FDFO desalination plant is to be used directly for fertigation, the nutrient

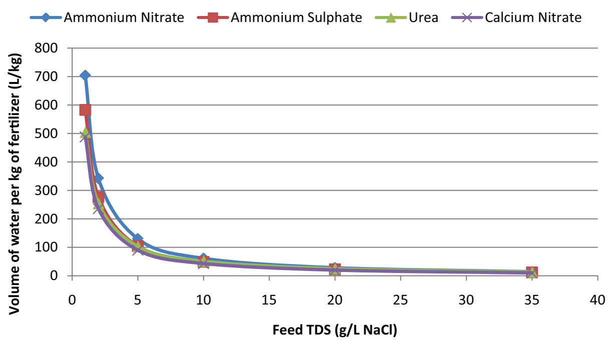

Fig. 10. Variation of water extraction capacities of the draw solutes by FO process at different feed TDS using different draw solutes.

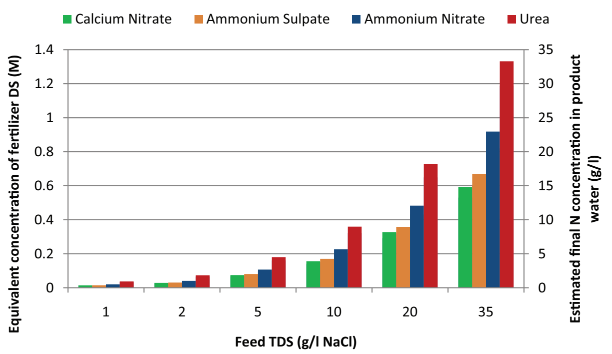

Fig. 11. Equivalent concentration of fertilizer DS and estimated final N concentration in product water for different feed TDS concentrations.

concentration must meet the water quality standards for irrigation. Therefore, it is important that the final FDFO produced water meets the nutrient concentration; otherwise, further dilution is required before applying for fertigation. Excessive fertilizer nutrient can be harmful to plants because it increases not only salinity but also toxicity [31]. In addition, leaching of fertilizer nutrients when excessive fertilizer is used in the water can cause undesired pollution of groundwater bodies [44].

Fig. 12 provides the highest recommended N concentrations for different types of plant crops. Plant requirement from nutrients varies depending on numerous factors, such as types of crop, cropping season, soil nutrient condition, etc. [45]. Generally, the required N nutrient concentrations ranges between 50 and 200 mg/L for N, function of the crop and growing time of year [46]. Comparing the information in Fig. 12 with that of Fig. 11, it can be easily concluded that it will not be possible to achieve the required water quality standards by the FDFO desalination process only, especially if feed salinity is more than 1 g/l. The N concentrations are significantly higher, especially for feed with higher TDS, indicating that a high dilution factor is needed to achieve recommended concentrations. This means that the additional dilution required is of several orders of magnitude before it can be used for direct fertigation.

For example, if the target crop is potatoes, being an important Egyptian crop, it is necessary for the N nutrient concentration to be 150 mg/L (Fig. 12). None of the four fertilizers achieve an acceptable N concentration for the potatoes without dilution before the fertilizer solution can be used for fertigation even with the lowest FS concentration of 1 g/l NaCl. Using the selected four fertilizers as the DS will require a dilution factor of at least 4 to make the N concentration acceptable for the potatoes at 150 mg/L using feed with TDS of 2 g/l. The dilution factor for $Ca(NO_3)_2$, SOA, NH_4NO_3 and urea are 4.8, 5.0, 6.8 and 12.2, respectively, when used with FS TDS of 2 g/L. As the FS TDS increases, the dilution factor will increase.

3. Fertilizer selection

In light of the above screening, ammonium sulphate was selected as the best draw solute for FDFO application in Egypt. The selection was based on the following justifications:

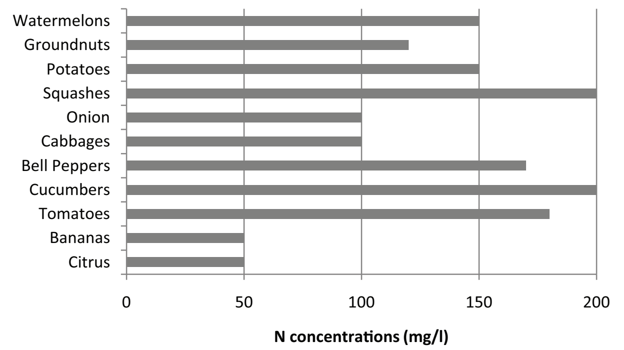

Fig. 12. Highest recommended N concentrations for different types of plant crops [46].

- Ammonium sulphate is the most non-expensive fertilizer, which will save operational costs (Fig. 4). It has been used in Egypt a long time ago, and it is produced locally by numerous fertilizer factories [33]. Although domestic demand for the granular ammonium sulphate is low, the crystal form is popular in Egypt since it is relatively cheap [47]. It is reported that Egyptian market consumed 140,000 ton of ammonium sulphate in 2012 [48].
- Ammonium sulphate produced osmotic pressure that is way higher than seawater (~28 atm) and brackish water, indicating its suitability to be used as an osmotic DS (Fig. 5).
- Ammonium sulphate provides the plant with nitrogen and sulphur at the same time as it contains approximately 21% nitrogen and 24% sulphur, promoting plant growth and crop yield. Because ammonium sulphate contains mainly ammonium nitrogen, it secures a lasting and sustainable nitrogen source. In the meantime, it minimizes nitrogen washing out from the soil. In addition, ammonium sulphate promotes the availability of secondary nutrients like manganese, iron and boron in the soil [31,45].
- Cost of ammonium sulphate is not affected by the fluctuating costs of natural gas because it is a byproduct of other industries such as steel and polyester compounds. Certain byproducts that contain ammonia or sulphuric acid are commonly converted to ammonium sulphate for use in agriculture [49,50].
- Ammonium sulphate is not hygroscopic (tendency to absorb moisture from the air), thus long storage duration is possible [51].
- Compared with urea, ammonium sulphate is more resistant to valorization.
- Ammonium sulphate is the preferred fertilizer for flood irrigation used for rice cultivation, while nitrate-based fertilizers are a bad option due to significant denitrification losses [51].
- Ammonium sulphate exhibits moderate final nitrogen concentration in product water so it can easily meet irrigation water quality (Fig. 11).
- Ammonium sulphate has SO_4^{2-} ionic species, which exhibit a large hydrated diameter compared with other fertilizer species. The effective diameter of the hydrated NH_4^+ and SO_4^{2-} ions are 250×10^{-12} and 400×10^{-12} m, respectively, making it hard to pass through the membrane material [29]. Consequently, ammonium sulphate is expected to perform well in terms of RSF [52].
- Ammonium sulphate is highly soluble in water, non-flammable and less hazardous than other draw solutes [50].

Other three fertilizers were overlooked due to the following reasons:

- Urea is not the best candidate as a DS. Not only because it exhibits the lowest osmotic pressure compared with other DS, but also because it results in the highest final nitrogen concentration in final product water, which will lead to need for dilution to meet water quality standards. In addition, other studies reported that urea suffers from significant reverse permeation of draw solutes compared with other DS. The high Reverse Solute Flux

(RSF)/ Specific Reverse Solute Flux (SRSF) of urea can be attributed to its low rejection by the membrane as urea is a neutral solute with the smallest molecular size in comparison with other DS [43].

- Ammonium nitrate is hygroscopic (tends to absorb moisture from the air), less effective for flood irrigation and prone to leaching after application [51].
- Calcium nitrate is hygroscopic and must be kept under air-tight storage conditions
- Ammonium nitrate and calcium nitrate are not easy to obtain as they are commercially banned being main constituents in explosives manufacturing.

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