Evaluation of polyvinylpyrrolidone as draw solute for desalination forward osmosis systems

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ABSTRACT

In this study, polyvinylpyrrolidone (PVP) was evaluated as a potential draw solute for desalination forward osmosis (FO) systems. The effect of various draw solute and draw solution parameters on the efficiency of FO operation was investigated, including PVP molecular weight, PVP concentration in solution, and the salinity of feed solution. Experiment results showed that at draw solution initial concentration of 200 g/L and feed solution initial salinity up to 15 g/L, the PVP-based draw solution can offer water flux up to 14.23 LMH in FO mode with raw material cost only at 0.61 USD/m³. PVP K17 was proven to be an effective draw solute for FO systems, providing good water flux and low reverse draw solute flux; while also being relatively non-toxic, non-corrosive, cheap and widely available compared to other types of novel draw solutes.

Key words: desalination, draw solute, forward osmosis, nano filtration, polyvinylpyrrolidone, water treatment

HIGHLIGHTS

• Novel draw solute for forward osmosis.
• Effective in desalinating low salinity water.
• Low price.
• Widely available.
• Non-toxic and non-corrosive.

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From ancient times, water has always been considered an important resource that played crucial roles in the rise and fall of human civilizations (Yevjevich 1992). For its essential and irreplaceable parts on the health, dignity, and prosperity of people, the access to water has been recognized by the United Nations as an important human right, and could significantly affect the realization of other fundamental human rights (UNGA 2010; OHCHR 2014). In modern times, with the challenges caused by rapid growth of human population and climate change, it has become increasingly clear that we must strive to ensure sufficient, safe, acceptable, physically accessible, and affordable water for personal and domestic use in order to avoid armed conflicts and minimizing human sufferings (Karnieli et al. 2019; Lu et al. 2019).

Owing to their ability to economically transform non-fresh water sources such as seawater, brackish water, or polluted inland water… into fresh water, desalination processes has been widely utilized by countries around the world as an important solution to fight the problem of water scarcity (Schiffler 2004; Islam et al. 2018). Out of numerous applicable desalination processes, forward osmosis (FO) has been considered as one of the most promising technologies for desalination, with the advantages of low energy cost in comparison to thermal processes, as well as high rejection of many pollutants and low membrane fouling in comparison to other membrane processes (Lutchmiah et al. 2014; Akther et al. 2015; Chekli et al. 2016).

Similar to other membrane desalination processes, fresh water is produced in FO systems by the difference in pressure on opposite sides of the semi-permeable membrane. In particular, the FO process is driven by the natural difference in osmotic pressure between one solution with lower osmotic pressure (the ‘feed solution’) and another solution with higher osmotic pressure (the ‘draw solution’), thus requiring minimal amount of input energy (Cath et al. 2006). Since osmotic pressure of feed solutions are generally fixed, the selection of draw solutions plays a pivotal role in all FO processes, and is considered one of the two main study subjects for most investigations involving forward osmosis desalination – beside the fabrication of semi-permeable membranes.

The selection of suitable draw solute (the ‘solute’ used to prepare draw solution) can greatly influence the efficiency and sustainability of FO operations. Ideally, the draw solute and/or its corresponding solution should meet the following six
requirements: (1) possess high osmotic pressure, (2) cause minimal reverse solute diffusion, (3) must be easy to regenerate from its diluted solution form, (4) be non-toxic, (5) be accessible and affordable, and (6) be non-corrosive to components of the FO systems. To date, various types of draw solutes have been explored, including but not limited to inorganic fertilizers (Phuntsho et al. 2011), ammonium bicarbonate (Kim et al. 2015), EDTA salts (Wang et al. 2019), phytate salts (Ge et al. 2018), magnetic nanoparticles (Na et al. 2014), or novel organic polymeric and supramolecular substances (Zhao et al. 2014; Wu et al. 2019).

In recent years, inorganic salts are still the go-to draw solute for most FO desalination systems due to their affordability and being easy to recover. The major drawbacks of these salts are that they can be quite corrosive to most conventional materials and can cause rapid membrane degradation, while also having high reverse solute diffusion. Thus, many studies were conducted to develop novel draw solutes with less corrosivity and lower reverse solute diffusion (Imane et al. 2019). While generally successful in achieving their stated objectives, those novel draw solutes are also less affordable and accessible, requiring complex synthesis processes, and in some cases must utilize rather complicated recovery methods. Therefore, it is important to continue searching for new draw solutes which more closely match the requirements for the ideal draw solute, so that future FO desalination systems could become more effective in producing fresh water.

Polyvinylpyrrolidone (PVP) is a polymer synthesized from the monomer N-vinylpyrrolidone. From previous literature, PVP was determined to be highly soluble in water, safe for human consumption as an approved food additive (E number E1201), widely available and affordable; while its aqueous solution was also generally non-corrosive to most metallic and other polymeric materials (Haaf et al. 1985; FAF 2020). In the field of forward osmosis, polyvinylpyrrolidone has been utilized in the fabrication and modification of the semi-permeable membrane (Morales-Torres et al. 2016; Wu et al. 2017), however there appears to be a lack of information on studies investigating the use of PVP as potential draw solute for FO systems.

In this study, the potential of PVP-based draw solution for FO desalination systems was determined through evaluating various operation parameters including water flux, reverse draw solute flux, draw solute recovery rate, effective water production rate, and effects of long-term exposure with draw solution on the forward osmosis membrane. The effects of draw solution concentration, feed solution salinity, and draw solute molecular weight on those operation parameters were also investigated, with stability and economic aspects being addressed by comparing with results from previous literature.

**METHODS**

**Materials**

PVP pellets (98%) of different molecular weights (PVP K15, PVP K17, and PVP K30) was purchased from Guangdong Yuemei Chemical (China). Natural sea salt was purchased locally in Vietnam. De-ionized water was freshly produced at the laboratory utilizing a Purelabflex-3 ultrapure water system (ELGA, UK).

Forward osmosis membranes were commercial cellulose-based FTSH2O forward osmosis membranes (Fluid Technology Solutions, US), while nano filtration membranes were TFC-based FilmTec™ NF90 nano filtration membranes purchased (DuPont, US). Both membranes were purchased from Sterlitech (US) in the form of flat-sheet 300 × 300 mm membranes, and cut into appropriate dimensions before use.

**Laboratory-scale forward osmosis experiment setup**

The setup of laboratory-scale forward osmosis – nano filtration experimental system is described in Figure 1. Membranes were cleansed with de-ionized water before/after each experiment. Before the first use and in-between experiments, membranes were immersed in de-ionized water to ensure water-saturation of their layers.

Forward osmosis experiments were conducted on an acetal copolymer CF042D-FO laboratory scale forward osmosis cell assembly (Sterlitech, US) with an active membrane area of 42 cm². All experiments were conducted with feed and draw solutions circulated concurrently into opposite sides of the forward osmosis cell assembly at average flow rates of 72 L/h, with the active layer of the membrane facing the feed solution (FO mode). The initial volumes for both feed and draw solutions was 5 L, so that the dilution effect would be minimized. The weight of the feed solution container was recorded every 15 minutes, for a total duration of 60 minutes. First recording of experimental data was conducted 15 seconds after turning on both pumps to ensure complete liquid filling and allow system stabilization.

Nano filtration (NF) experiments were conducted on an acetal copolymer CF042D laboratory scale crossflow cell assembly (Sterlitech, US) with an active membrane area of 42 cm². The assembly was equipped with a 1.75 mm thick stainless-steel spacer (Sterlitech, US). Experiments were proceeded at feed channel hydraulic pressures of 20 bar and feed channel average
flow rates of 240 L/h, so that diluted draw solution can be re-concentrated back to its original concentration. Theoretical draw solution recovery rate and effective water production rate were determined based on the amount of fresh water produced and the amount of draw solute remained in the recovered draw solutions.

The different parameters being investigated were concentration of draw solution, salt concentration of feed solution, and molecular weight of the draw solute. Each experiment was conducted in triplicate, with results averaged to minimize possible errors.

Effects of continuous operations on semi-permeable FO membranes

Experiments were conducted similarly as above, however the initial volumes for both feed and draw solutions was increased to 25 L, and the total duration of each experiment was extended to 5 hours. After each experiment, the pumps were momentarily stopped and replaced with new ones, while containers of both feed and draw solutions were also replaced with containers containing fresh feed and draw solutions, respectively. As such, the experiments could be considered as one continuous process, with the concentration of both feed and draw solutions maintained around suitable values.

The total continuous run-time for each set of these experiments was 100 hours, with data recording conducted every 20 hours to investigate the effects of long-term exposures with PVP-based draw solution on semi-permeable properties of the membrane. These effects were determined based on the changes in operation duration on water flux and specific operation draw solute loss throughout the operation duration. Each set of experiments was conducted in triplicate, with results averaged to minimize possible errors.

Additionally, the changes in microscopic structures of the semi-permeable membrane after 100 hours of operation duration were also observed by scanning electron microscopy (SEM) utilizing a Jeol SM-6510LV (Jeol, Japan) microscope at an accelerating voltage of 2.0 kV.
RESULTS AND DISCUSSION

The effects of PVP molecular weight

As described, PVP is a polymer synthesized from the monomer N-vinylpyrrolidone, with the molecular weight distribution range of each particular sample determined by its degree of polymerization. Commercially, the K-value of PVP is an index utilized to represent the molecular weight distribution range of one particular PVP product (Swei & Talbot 2003). Thus, commercial PVP products of different K-value were used in determining the effects of PVP molecular weight on FO operation.

Initial experiments suggested that PVP K60 (Mw = 270,000–400,000) and PVP K90 (Mw = 1,000,000–1,500,000) were both not particularly well-soluble in water, while their aqueous solutions at high concentrations were also very viscous. Due to challenges that arose while running these solutions through the FO system for an extended period of time, both PVP K60 and PVP K90 were not considered in subsequent experiments.

Studies on the effects of PVP molecular weight were conducted using aqueous solutions of PVP K15 (Mw = 8,000–12,000), PVP K17 (Mw = 10,000–16,000), and PVP K30 (Mw = 45,000–58,000) as draw solutions. In all experiments, feed solutions were de-ionized water, while the draw solutions were investigated at two solute concentrations of 50 g/L and 150 g/L.

Experiment results in Figure 2 suggested that at the draw solution concentration of 50 g/L, all three types of PVP showed potential as draw solutes with water permeation values after 60 minutes ranging from 12.2 mL to 16.2 mL. At the draw solution concentration of 150 g/L, water permeation values after 60 minutes increased to the range from 28.9 mL to 48.0 mL.

Interestingly, in all experiments, water permeation values showed a linear relationship with time. This can be explained by the initial amount of both feed and draw solutions utilized. Since the initial amount of both solutions was high, the volume of mass transfer through the semi-permeable membrane was relatively low, leading to negatable changes of concentration in both solutions. Thus, all mass transfer processes were stable throughout the 60-minutes duration of experiments, leading to constant increases in water permeation.

The results also suggested that draw solutions containing PVP K15 and PVP K17 exhibited similar forward osmosis potential, proven by the similarity in values of water permeation after 60 minutes exhibited at both draw solution concentrations of 50 g/L and 150 g/L. Additionally, these draw solutions also achieved markedly higher values of water permeation compared to that containing PVP K30, showing water permeation 1.3 and 1.5 times higher at draw solution concentrations of 50 g/L and 150 g/L, respectively.

These results can be explained by the differences in molecular weight range between PVP K15, PVP K17, and PVP K30. As described above, both PVP K15 and PVP K17 have comparatively lower molecular weight, and overlapping ranges of molecular weight distribution. Therefore, it is un-surprising that the solutions of these products can exhibit similar properties, including osmosis properties.

In contrast, PVP K30 has markedly higher molecular weight, thus having a longer and more bulky molecular structure. This structure promotes intra- and inter-molecule interactions between PVP K30 molecules, while reducing interactions between PVP K30 and water molecules. This leads to the decrease in osmosis potential, thus leading to the decrease in water permeation.

Figure 2 | Effects of PVP molecular weight on water flux at draw solute concentrations of 50 g/L (left), and 150 g/L (right).
permeation. Additionally, PVP products of higher K-values also exhibit higher viscosity, causing higher internal concentration polarization in the porous structure of semi-permeable membrane, which can negatively affect the mass transfer of water through the semi-permeable membrane.

Therefore, only PVP K15 and PVP K17 were considered for further investigations.

**The effects of PVP concentration**

Experimental results on the effects of PVP concentration on FO operations are shown in Figure 3.

The results showed that in the draw solution concentration range of 50–250 g/L, the solutions of both PVP K15 and PVP K17 exhibited similar water flux profiles, while the reverse draw solute fluxes of PVP K15 solutions were increasingly higher than that of PVP K17 solutions.

The difference in reverse draw solute flux of these solutions could be explained by the difference in molecular weight of PVP K15 and PVP K17. As described above, while sharing the same molecular structure, PVP K15 (Mw = 8,000–12,000) has noticeably smaller molecular size compared to PVP K17 (Mw = 10,000–16,000). Therefore, PVP K15 molecules can more easily travel through the porous structure of the semi-permeable membrane compared to PVP K17 molecules, leading to larger reverse draw solute flux.

It is also interesting to note that for both types of draw solutions, reverse draw solute flux increased linearly up until draw solution concentration of 250 g/L, while water flux only increased linearly up until draw solution concentration of 200 g/L. This was because at higher draw solution concentrations, viscosity of draw solutions also increased, causing higher internal concentration polarization in the porous structure of semi-permeable membrane, leading to reduced mass transfer of both water and draw solute. However, the mass transfer of water was at a much larger rate than that of draw solute at the same draw solution concentration, thus the results from such reduction in mass transfer was markedly greater for water than for draw solute.

Further observations illustrated in Figure 4 showed that with the increase in draw solution concentration, the ratio between water fluxes (Jw) and reverse draw solute fluxes (Js) also increased, especially for PVP K17. This means that for the same amount of fresh water produced, draw solute loss was higher for PVP K17 than for PVP K15, making PVP K15 the better draw solute candidate based solely on economic standpoint – since both products have similar price per unit of weight.

Based on these results, it is evident that PVP K17 was the better choice compared to PVP K15. Thus, only PVP K17 was selected to be used in further investigations.

**The effectiveness of NF regeneration**

Experiments were conducted using de-ionized water as feed solution, and aqueous PVP K17 solutions with concentrations ranging from 50 to 250 g/L as the draw solution. Experimental results are presented in Figure 5.

The results showed that both recovery rate and specific water flux of the NF system would decrease with the increase of initial draw solution concentration. However, at PVP concentrations lower than 250 g/L, all recovery rates remained

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**Figure 3** | Effects of polyvinylpyrrolidone concentration on water flux (left), and reverse draw solute flux (right).
above 98.5%/L. This could be explained by the fact that PVP polyvinylpyrrolidone was a polymeric substance with long-chain molecular structure, thus it would be harder for polyvinylpyrrolidone molecules to pass through the NF membrane comparing to other significantly smaller inorganic substances.

The decrease in specific water flux can be attributed to the decrease in the driving force of the NF process caused by a reduction in the difference between osmotic pressure of diluted draw solution, which would naturally increase when increasing initial draw solute concentration, and the fixed hydraulic pressure generated in the NF system. In this case, the specific water flux of NF system was satisfactorily high, and could be configured with reasonable boundaries of hydraulic pressure to create one continuous FO-NF system.

In combination with previous results, the concentration of draw solution based on PVP K17 was determined to be 200 g/L, and this solution concentration shall be used in further investigations.

The effects of feed solution salinity

Experiments were conducted under previously stated parameters, however the feed solution for FO system was replaced with aqueous solutions of sea salt dissolving in deionized water, and with feed solution concentrations ranging from 5 to 25 g/L. Experimental results are presented in Figure 6.

Per experimental results, the 200 g/L aqueous solution of PVP K17 was proven effective in drawing fresh water from saline feed solution with salinity up to 15 g/L. Beyond such salinity threshold of feed solution, reverse water flux was observed.
The reverse in water flux at higher feed water salinity was expected, and can be easily explained by the fact that increasing the feed water salinity would also increase its natural osmotic pressure. With osmotic pressure of the draw solution remaining the same, such increasement in osmotic pressure of the feed solution would change the difference in osmotic pressure between two sides of the semi-permeate membrane.

In this case, the positive osmotic pressure difference between the draw solutions and the feed solutions would be reduced with the increase in feed solution salinity, thus reducing positive water flux. Once the osmotic pressure difference became negative at feed solution salinity of 20 g/L, a reversion in water flux was observed, with fresh water being drawn from the draw solution side to the feed solution side.

Membrane stability with continuous operations and economical assessment

Experiments were conducted with de-ionized water as the feed solutions, and 200 g/L PVP K17 aqueous solution as the draw solutions.

Results presented in Figure 7 showed that, with extended continuous FO operation up to 100 hours, there was negligible deterioration of the semi-permeable membrane caused by PVP K17. Water flux was only slightly reduced by 0.98% after 100 hours of continuous operation, from 14.23 LMH to 14.09 LMH. Interestingly, the water flux recorded of membranes after 20-hours operation was higher than that of new and unused membranes, with water flux increased from 14.23 LMH to 14.25 LMH.

Figure 7 | Effects of operation duration on water flux (left), and specific operation draw solute loss (right).
These results can be explained by changes in the structure of semi-permeate membrane, as shown in Figure 8. SEM image of the unused membrane (left) showed a non-continuous surface, combined of smooth areas and ‘bubbles-like’ areas – which was potentially caused by the production method of such membrane. However, SEM image of the membrane after 100-hours continuous operation (right) showed a smoother surface, with minor solid particles and aggregates deposited on the surface.

The changes in membrane surface were probably caused by the affinity of PVP with the composition of FO membrane, which was proven by its extensive uses in FO membrane fabrication and modification as shown in literature (Morales-Torres et al. 2016; Wu et al. 2017). Thus, continuous exposure to PVP solution helped to make the upper layer of FO membrane become more uniform with a smoother surface, while also slightly increasing the effectiveness of the membrane as shown by the increase in water flux after 20-hours continuous operation. The upper layer of FO membrane remained intact, since the dissolve of this layer would expose the highly porous supporting layer below.

However, with increased continuous operation, solid contents originally dissolved in the feed solution or later introduced by components of the FO system would continue to deposit on the membrane surface, obstructing the mass transfer of water through the semi-permeate membrane. Thus, there was a reduction in water flux after more than 20 hours of continuous operation.

The economic efficiency assessment of PVP K17 draw solute comparing to existing works was showed in Table 1. This assessment proved that the FO operating cost of PVP K17 draw solute was rather low, making it an economical and safe potential draw solute for forward osmosis systems.

**CONCLUSIONS**

The results obtained from laboratory-scale FO and NF experiments suggest that PVP could be utilized as potential draw solutes for hybrid FO/NF processes, especially the commercially available product PVP K-17. Good water fluxes could be achieved with draw solution concentration of 200 g/L and feed solution salinity up to 15 g/L. The solute rejection was 99.21% for FO desalination processes and 99.01% for NF regeneration process, which combined to an operation cost for the draw solute of only 0.61 USD/m^3, lower than both other traditional inorganic salt-based draw solutes and novel organic

![Figure 8](image_url) The surface of unused FO membrane (left), and that of FO membrane after 100 hours of continuous operation.

**Table 1** The economic efficiency of PVP solute, comparing to existing works (Gwak et al. 2015)*

| Draw solute | \( C_{\text{DS}} \) g/L | Cost, S/kg | Cost, S/L | \( J_s/J_w \), g/L | FO operating cost, USD/m^3 |
|-------------|-----------------|--------|--------|----------------|----------------------------|
| NaCl        | 35.2            | 1.1    | 0.04   | 0.74           | 0.81                       |
| MgCl\(_2\)  | 33.8            | 6.2    | 0.21   | 0.57           | 3.53                       |
| PAspNa      | 200.0           | 8.8    | 1.76   | 0.19           | 1.67                       |
| PVP K17     | 200.0           | 5.5    | 1.10   | 0.11           | 0.61                       |

*All costs were re-adjusted to match that of Vietnam domestic market prices.
substance-based draw solutes. In conclusion, PVP K17 was proven to be a promising draw solute for hybrid FO/NF processes, especially in developing countries being challenged with the salinization of their water sources.

However, it should also be noted that this study is only a preliminary research, and further works need to be done in order to explore unforeseen challenges and optimize the operational parameters of this proposed process. The economic and membrane stability aspects should also be further explored, since laboratory-scale experiments cannot reflect the true operational costs and membrane wearing occurred in pilot-scale and industrial-scale operations.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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