Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2017.20680

77 (2017) 129–134
May

Evaluation of EDTA salts for potential application to draw solute of a forward osmosis process

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Received 6 October 2016; Accepted 16 December 2016

ABSTRACT

The selection of novel draw solution is one of the critical research fields in a forward osmosis (FO) process. Among various materials, ethylenediaminetetraacetic acid (EDTA) was suggested as a good candidate for draw solution due to its high solubility and easy recovery. However, it is important to find the optimum pH of EDTA in the FO process since EDTA form can be changed according to pH values. This study was initiated to find the optimal pH value of the EDTA salt used as a draw solution, as well as to explore the possibility to recover it by using the nanofiltration (NF) process. From the results of this study, the pH value that has the lowest amount of reverse salt flux (RSF) of EDTA and Na^+ ions was found to be 7. In case of the rejection rate test of the NF process, two different membranes, each made with different materials, were used (cellulose acetate [CA] membrane and polyamide [PA] membrane). Both of the membranes have high rejection rate, which is over 99.15% at low pressure (5 bar). From the result of water flux, the performance of PA membrane was about twice as good as the CA membrane under all pressures (5, 10, 15 bar). It could be concluded that the EDTA salt has high water flux with low RSF in the FO process as well as the high salt rejection rate in the NF process. In addition, use of the PA membrane is much preferred than the CA membrane for EDTA salt recovery in the NF process.

Keywords: Forward osmosis; Draw solution; EDTA; Nanofiltration; Recovery of draw solution

1. Introduction

Sustainability in terms of water, energy, environment and climate is the rising concerns of the century, as a result of rapid growth in population, economics and industrialization [1,2]. Numerous communities are already affected by clean water scarcity, energy crisis, environmental pollution and climate change [3–5]. Fortunately, a host of technologies are developing in order to secure abundance and steadiness of freshwater such as seawater desalination, water recycling and reuse, treatment of low-quality local water and capturing rainwater [6]. Forward osmosis (FO) has proven to be quite promising with its excellent advantages such as high rejection of many pollutants, without the aid pressure and low membrane fouling [7–9].

In the FO process, the semi-permeable membrane serves as a separation medium, through which the osmotic pressure difference allows the clean water to be permeated from the feed solution (FS) to draw solution (DS). During this process, the semi-permeable membrane rejects other ions or molecules in the FS [3,10,11]. The FO process may seem advantageous, yet faces some critical challenges: membrane materials, membrane concentration polarization, membrane fouling and reverse solute diffusion [12]. Hence, developments of new membranes and novel draw solute for suitability in FO process are highly encouraged. The majority of the study worked in the literature mentions the development of new membranes and process designs [13–15]. Yet the study in search of the novel draw solute has been barely attended to, for future FO process applications.

In order for the advancement of FO technology, the availability of a suitable DS is critical. The characteristics of

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an ideal draw solute, fundamentally, contain high osmotic efficiency, high solubility in water and minimal reverse draw solute flux. It should also be separated at a low cost and effort to yield potable water [3,16]. Currently, the most widely used draw solute is sodium chloride (NaCl). NaCl has high water flux along with the RO process as its recovery method. RO process has high selectivity for salt and successfully produces high quality water, yet it consumes large amounts of energy along with its expensive equipment and high fouling propensity [17,18]. In fact, NaCl also has a high reverse salt flux (RSF). The exploration of suitable DSs for FO process dates back to many effortful decades [3]. Substitutes have been reported, which include ammonium bicarbonate (NH_4HCO_3) from ammonia and carbon dioxide, magnetic nanoparticles (MNP), glucose, etc. The recovery method of ammonium bicarbonate is heating at temperatures as low as 40°C–60°C. Although the system is incredibly competitive, the RSF of monovalent ions (NH_4^+) still proved to be problematic [19]. MNP was also captured by a canister separator for recovery. Poor performance and agglomeration were its problems [20,21]. Glucose as a DS for the FO process also has a low flux and does not produce pure water [22,23].

Several organic compounds were used as DSs; ethylenediaminetetraacetic acid (EDTA) disodium was studied in FO-nanofiltration (FO-NF) process as a DS. It has high water solubility, high water flux and nontoxic materials. Therefore, it is good for using as draw solute. EDTA has a different form according to pH values. EDTA disodium salt allows good solubility at a high pH value, which provides the trivalent and tetravalent ions to remain at the draw side of the solution much easily, limiting the diffusion of counter-ion [24]. EDTA as a DS in terms of FO performance was successful, and it was recovered through the NF membrane with a high rejection rate. Furthermore, because EDTA sodium salt is environmentally friendly, it can be stated that EDTA salt is a reasonable DS [25]. And pH value of EDTA is an important factor in FO process due to formation of EDTA.

Therefore, this study evaluates the water flux and RSF according to the pH values and the rejection rate of EDTA salt in the NF process. The conformity assessment of EDTA salts as DSs for FO-NF process are to be assessed through the two experiments.

2. Materials and methods

2.1. Characterization of draw and feed solution

Laboratory-grade EDTA-2Na (purity of 99%) was purchased from Sigma-Aldrich Co. (Germany) and used as DSs. NaOH (purity of 98%) and $Ca(OH)_2$ (purity of 95%) used to adjust pH from 5 to 10. The pH values of DSs were measured by a pH meter (Thermo scientific, USA). 0.3 M DS of 500 mL adjusted at target pH was made by using above materials and used in FO filtration test. Osmolality of DS was measured by a freezing point osmometer (Fisk® Micro-Osmometer Model 210, USA) to analysis DS's osmotic pressures, which are different according to target pH at the same DS concentration. Deionized (DI) water of 1 L was used for FS for all FO filtration test.

2.2. FO process

Commercially available flat-sheet thin film composite FO membrane was provided by Porifera, Inc. (USA) and used for all FO experiments. This membrane's active layer is believed to be manufactured by polyamide (PA), which is supported by polysulfone and polyester materials [1]. Fig. 1 shows schematic illustration of FO filtration test. Lab-scale cross-flow filtration system was set up for FO filtration test. The custom-built flat and frame cell was fabricated to make FO membrane locate in it. The cell consisted of two symmetric channel and which dimensions of 77 mm long by 26 mm wide by 3 mm deep (membrane effective area: 20.02 cm^2). All FO filtration test was progressed in FO mode (active layer faced the feed side). No spacer on both of sides was used in this experiment. The FS and DS's flow rates (cross-flow velocity: 8.55 m/cm) were steadily controlled by gear pumps (Longer Pump WT3000-1FA), which was calibrated before filtration test for prevention from flow rate change. In all these tests, the pressure difference between DS and FS was maintained under 0.1 bar to minimize hydraulic pressure effect on water flux. And also to fully saturate FO membrane, FO membrane was cross-filtrated by DI water before FO filtration test. A chiller (CPT Inc., Korea) was used to keep each tank constant at target temperature (25°C), and the fluctuation of temperature was under 1°C. A digital balance (AND GF-6000, USA) located under the DS tank was connected to computer to check volume of DS and calculate water flux. Water flux of initial 15 min was discarded for water flux stabilizing. Filtration test progressed until 100 mL of DS migrated into feed side.

2.3. NF process

To compare membrane performance for regeneration of draw solute according to membranes, two commercial NF membranes were used for the NF test (regeneration DS; Table 1). Prior to the filtration test, membrane was soaked in DI water over 24 h and filtrated with DI water at 20 bar until a constant value of flux was obtained.

Fig. 2 shows the NF filtration test schematic. Likewise, FO filtration test, lab-scale cross-flow filtration system was set up using custom-built NF cell for NF test. The cell consisted of two symmetric channel and which dimensions of 190 mm long by 140 mm wide (membrane effective area: 128.35 cm^2), and 34 mil spacer was used for NF test.

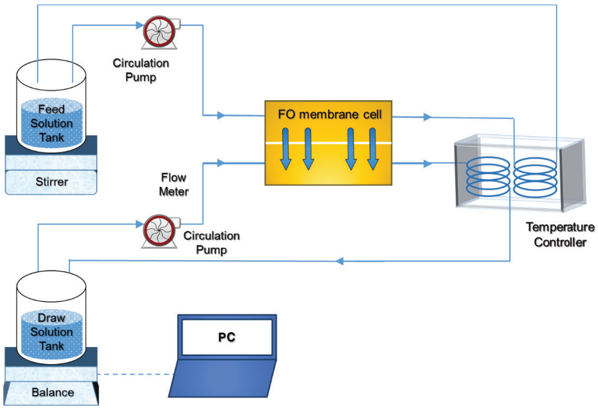

Fig. 1. Schematic illustration of FO filtration test.

Table 1
List of NF membranes used for the NF test

Membrane type	Model	MWCO	NaCl rejection	$MgSO_4$ rejection
Polyamide (PA)	TS80	150 Dalton	80%–90%	98%
Cellulose acetate (CA)	SB90	150 Dalton	85%	98%

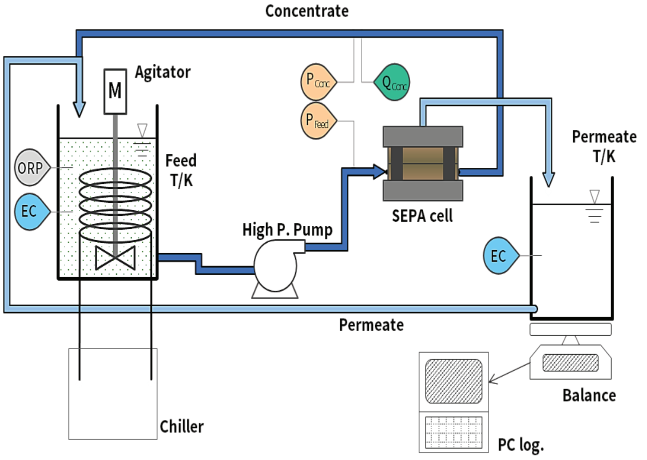

Fig. 2. Schematic illustration of the NF test.

Unless otherwise defined separately, the following operation condition was applied for NF test. EDTA slats with concentration of 0.03 M was conducted at operating pressure of 5, 10 and 15 bar, respectively, feed flow rate of 3 LPM, and operating temperature of 25°C. 15 min of initial water flux was neglected for water flux stabilizing in each test.

2.4. Measurement of flux and rejection rate

The water and RSF in FO filtration test were calculated by the following equations:

$$J_w = \frac{\Delta V}{At} \tag{1}$$

$$J_s = \frac{C_t V_t - C_0 V_0}{At} \tag{2}$$

where J_w is the water flux of a membrane ($L/m^2/h$, abbreviated LMH), and J_s means the RSF of a membrane ($mmol/m^2/h$). V_t is the volume of draw tank at time t; V_0 is the volume of draw tank at time 0; C_t is the concentration of the draw solute in the feed tank at time t; and C_0 is the concentration of the draw solute in the feed tank at time 0. Also, A means the membrane area (m^2), and t is the operating time.

For classification of RSF in FS of FO, ion chromatograph and total organic carbon (TOC) were used to measure Na, Ca and EDTA using predetermined calibration curve, respectively.

The water flux and rejection rate in NF filtration test were calculated by the following equations:

$$J_w = \frac{\Delta V}{At} \tag{3}$$

$$R = \left(1 - \frac{C_p}{C_f}\right) \tag{4}$$

where J_w is the water flux of a membrane (LMH); R is the rejection rate of a membrane (%); ΔV is the change volume of permeate. C_p is the concentration of permeate at time t, and C_f is the concentration of the FS at time 0. Also, A means the membrane area (m^2), and t is the operating time.

For calculation rejection rate of draw solute in FS of NF, TOC was used to measure EDTA using predetermined calibration curve.

3. Results and discussion

3.1. Possibility evaluation using EDTA salts as draw solution in FO process

3.1.1. Water flux according to pH values of draw solution in FO

EDTA form is changed by pH. EDTA is adsorbed as a binuclear complex at low pH and as a mononuclear complex at high pH [26]. Therefore, pH value is important parameter for EDTA. Osmolality and water flux according to DS's pH were determined from lab-scale FO setup. The experiment of FO process is conducted in the FO mode with 0.3 M EDTA-2Na solution as DS and DI water as FS. Fig. 3 shows the osmolality and water flux for different pH values. In case where pH 5 was adjusted by $Ca(OH)_2$, it was not possible to raise the pH of DS above pH 5 because the solubility of $Ca(OH)_2$ is much lower than the solubility of NaOH [27]. In both cases for adjusting the pH, using $Ca(OH)_2$ and NaOH, osmolality increased by the level of DS's pH due to injection of pH adjustment solution. Water flux had a proportional relation with osmolality, and this is general results that are commonly known in other literature [19]. Although NaOH is a good choice of alkali to increase the pH of EDTA salt solution, using NaOH to obtain a high pH would result in an increase of the concentration of Na^+ and OH^- ion, which, in turn, increase the osmolality [28]. Therefore, increase of pH would result in high osmolality and, consequently, a higher osmotic pressure. From pH 5 to 10, osmolality values are 728, 813, 944, 992, 1,022 and 1,030 Osmol/kg, respectively, and water flux values were 7.16, 8.46, 10.34, 11.08, 11.75 and 11.08 LMH, respectively. An increase in the pH of DS above the isoelectric point of the PA membrane surface, which usually lies in the range of pH 3–9, resulted in an increased negative charge [29]. The high negative charge of the FO membrane at high pH values results in increase in water flux [30]. However, though increase by 8 Osmol/kg from pH 9 to 10 of DS, there was water flux reduction by 0.7 LMH. In this respect, we can find that external concentration polarizations phenomenon could be important factor to determine optimal condition of FO-RO process using EDTA as DS.

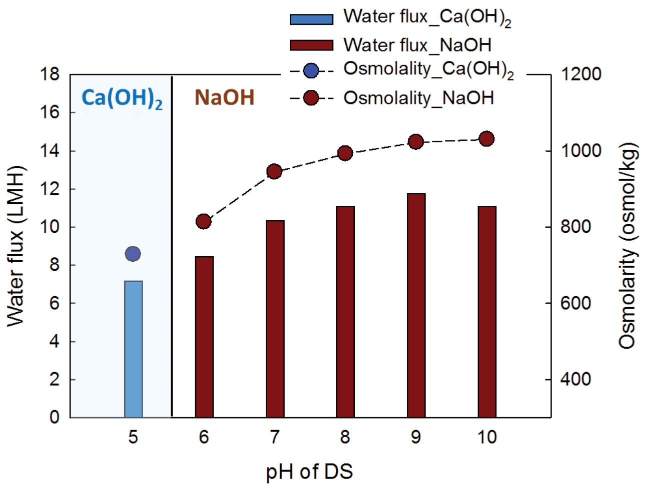

Fig. 3. Osmolality and water flux adjusted by $Ca(OH)_2$ and NaOH.

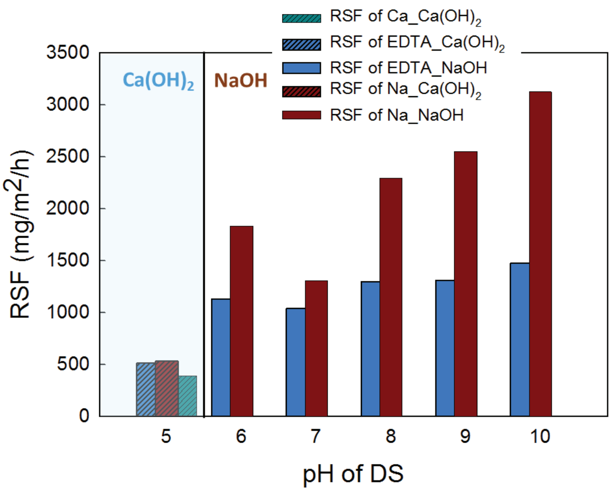

Fig. 4. RSF variation according to pH values of draw solution.

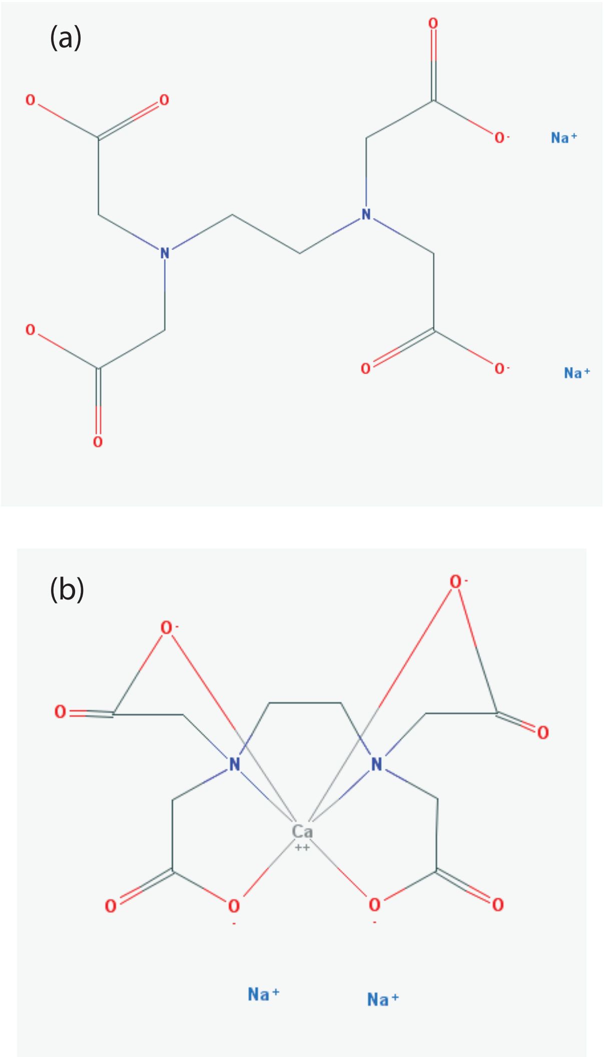

Fig. 5. Structures of metal-EDTA complexes and EDTA disodium salt. (a) EDTA disodium salt and (b) EDTA calcium complex.

3.1.2. Reverse salt flux (RSF) according to pH values of draw solution in FO

Interestingly, the RSF-EDTA value was significantly lower for the case where pH was adjusted by $Ca(OH)_2$, than the case where pH was adjusted by NaOH (Fig. 4). This could be attributed to chelating compounds, combined by EDTA and Ca^{2+}, forming a bigger molecule in size. This increased molecular size makes it difficult to pass through the FO membrane for EDTA. As can be seen from Fig. 5, the structure of EDTA-2Na and EDTA-Ca complex has a different molecular size. EDTA-Ca complex is bigger than EDTA-2Na formation.

There were RSF-EDTA reductions as the pH of DS increased from 6 to 7. However, when the pH was over 8, both RSF-Na and RSF-EDTA increased again. At pH 6, the dominant EDTA complex ion is H_5-$EDTA^+$. Other complexes that exist include H-$EDTA^{3-}$. At this pH, the cations attract closer to the membrane. Because FS has a lower concentration of these ions as compared with DS, some of these molecules can diffuse to the feed side because of the concentration gradient. However, at pH 7, the ratio of H-$EDTA^{3-}$ increases. This prevents EDTA from easily passing through FO membrane because of repulsion force between membrane and H-$EDTA^{3-}$ [25]. At pH 8, the predominant composition of H-$EDTA^{3-}$ is higher than other complexes. And at pH 9 and 10, Na-$EDTA^{3-}$ is the most abundant complex in the EDTA solution. However, from pH 8 to 10, the RSF is higher than that of pH 7. It is thought that the ionic strength makes membrane surface to neutralization [25]. Bunt et al. [31] found that charge and hydrophobicity of the cell surface is affected by pH and ionic strength [32]. As the pH increases, the NaOH used for adjusting pH also increases, which in turn means the concentration of the Na^+ ion increases. Consequently, Na^+ ion attaches to the membrane surface, and this makes the membrane surface to be neutral. Because of the neutral membrane surface, the repulsion between the Na-$EDTA^{3-}$ or H-$EDTA^{3-}$ and the membrane surface decreases. Therefore, the RSF increases since the EDTA can diffuse through the membrane more easily [33]. And high level of RSF could influence water flux at high DS's pH. Therefore, the optimum pH is pH 7 in this process.

3.2. Recovery using EDTA salts as draw solution in NF process

3.2.1. Water flux of diluted draw solution in NF

The second part of this study conducted using a lab-scale NF test to determine the regeneration of DS and evaluate the quality of permeate. Two NF membranes composed of cellulose acetate (CA) blend and PA materials and diluted DS adjusted to pH 7 using NaOH were used for NF filtration test. Concentration of diluted solution is 0.03 M, and this one-tenth of the concentration of the diluted solution was used in FO process, which is 0.3 M. Fig. 6 shows the comparison of water flux at three different pressures: 5, 10 and 15 bar, using different membrane materials. At the three different pressures, water flux values were 11.42, 25.20 and 43.04 LMH with the use of CA blend membrane and 27.68, 70.74 and 116.81 LMH with the use of PA membrane, respectively. This is somewhat obvious, since increase in operating pressure would result in the increase in water flux. And PA membrane has a higher water flux values compared with CA blend membrane. Therefore, PA membrane is preferable membrane than CA blended membrane for EDTA recovery system.

3.2.2. Rejection rate of diluted draw solution in NF

Rejection rate of DS is one of the most important factors in FO-NF process. The rejection rate is important, because the possibility of draw solute recovery can be determined according to the rejection rate of the EDTA salts. Also, if the draw solute has a high rejection rate, it can be used as DS. Therefore, the experiment for the rejection rate of EDTA salts in lab-scale NF test was done for this study. Fig. 7 shows the comparison of rejection rate of EDTA at three different pressures, 5, 10 and 15 bar, using different membrane materials. With the CA blend membrane, the rejection rates were 9.44%, 99.44% and 99.61%, and 99.15%, 99.37% and 99.59% for the PA membrane at 5, 10 and 15 bars, respectively. This proves that the higher operating pressure results in a higher rejection rate. Also, from the results shown in Fig. 7, it can be concluded that using the CA blend membrane has relatively higher rejection rate than the PA membrane. However, the difference was minimal and can be considered negligible since both EDTA rejection rates were over 99.15%. Therefore, the NF recovery of EDTA salts indicated that all NF membrane performed well.

4. Conclusions

This study evaluates the water flux and RSF according to the pH values and the rejection rate of EDTA salt in the NF process. The conformity assessment of EDTA salts as DSs for FO-NF process is to be assessed through the two experiments. This study demonstrates finding optimum pH value and possibility of recovery using the EDTA salt as DS. The results obtained from laboratory-scale FO and NF test suggest that EDTA salt could be a potential draw solutes for hybrid FO-NF process. In FO process using EDTA salts as DS, the water flux was about 10 LMH, and the amount of RSF of EDTA was low under 1,000 mg/m^2/h. Due to repulsion forces between the EDTA and the FO membrane, there were RSF-EDTA reductions as the pH of DS increased from 6 to 7. Therefore, the optimum pH is 7 in FO process. In NF process, PA membrane had a higher water flux than CA membrane. Also, due to the rejection rate of EDTA over the 99.15%, it is possible to recover EDTA at a low pressure (5 bar) using high water permeability NF membrane. Thus, in this work, EDTA salts have possibility as a draw solute in FO-NF process. However, further studies are needed to overcome the limitation of low solubility using EDTA. Hence, the next step is to compare the EDTA and the EDTA complex as DS.

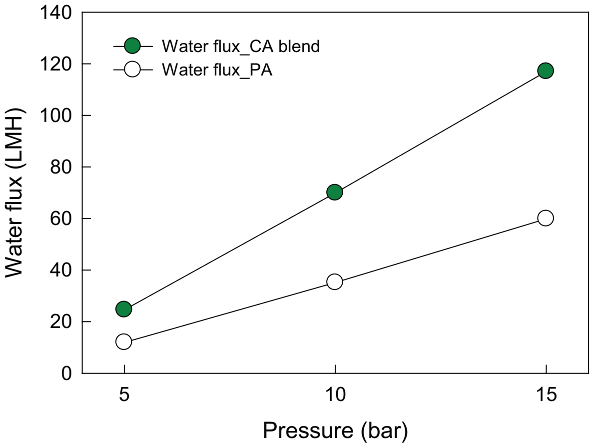

Fig. 6. Water flux using diluted draw solution adjusted by NaOH at pH 7.

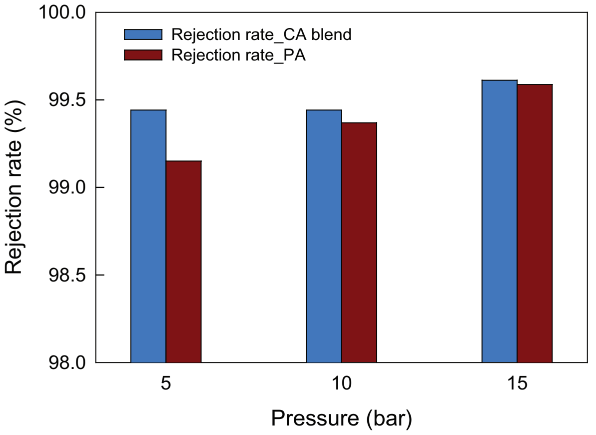

Fig. 7. Rejection rate using diluted draw solution adjusted by NaOH at pH 7.

Acknowledgment

This research was in part supported by a grant (code 16IFIP-B088091-03) from Industrial Facilities & Infrastructure Research Program funded by Ministry of Land, Infrastructure and Transport of Korean government.

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