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Tandem Desalination/Salination Strategies Enabling the Use of Redox Couples for Efficient and Sustainable Electrochemical Desalination

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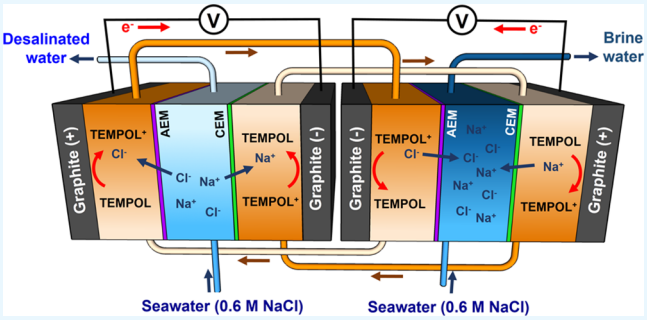
ABSTRACT: As access to fresh water becomes an increasingly serious global issue, developing desalination methods that can reduce not only the cost but also the carbon footprint of desalination has become of utmost importance. In this study, we demonstrate the use of the oxidation and reduction of the same redox couple with fast redox kinetics as the anode and cathode reactions of an electrodialysis (ED) cell. This reduces the thermodynamic equilibrium cell potential to 0 V while also significantly reducing the kinetic overpotentials required for cell operation. As a result, the overall operating voltage of our ED cell is remarkably reduced, making it possible to use ED for seawater desalination and to operate the ED cell by using inexpensive portable power generators that provide a limited voltage. The sustainable use of the redox couple in the ED cell was enabled by a new strategy, where a desalination ED cell and a salination ED cell were operated in tandem. In this tandem system, the electrolytes in the anode and cathode compartments of the two cells were circulated such that the compositional changes of the electrolytes made in the desalination cell could be reversed in the salination cell. As a result, the feedwater (0.6 M NaCl) could be converted to 0 and 1.2 M NaCl solutions in the desalination cell and salination cell, respectively, without the accumulation of salt ions in the anode and cathode compartments. The operating principles and performance of a proof-of-concept tandem desalination/salination system are demonstrated.



KEYWORDS: *desalination, electrodialysis, seawater desalination, TEMPOL, tandem electrodialysis, salination*

1. INTRODUCTION

The lack of access to fresh water is one of the most serious issues that the world is facing.[1−3] Considering the abundance of seawater on earth, seawater desalination has the possibility to offer a sustainable and unlimited supply of fresh water.[4−11] To date, distillation and reverse osmosis (RO) have been primarily used for seawater desalination with RO being the more cost-effective option.[11−14] However, the cost for RO is still high due to the considerable electrical energy input required to operate high-pressure pumps.[12−16]

Another desalination technology is electrodialysis (ED), which has been used for brackish water desalination.[17−22] A scheme of an ED cell composed of three compartments is shown in Figure 1A. Oxidation occurs in the anode compartment of the cell, resulting in the consumption of anions (e.g., via Cl^- oxidation), and reduction occurs in the cathode compartment, resulting in the consumption of cations (e.g., via proton reduction). To maintain charge neutrality, Cl^- from the middle compartment, which contains a saline solution, will move toward the anode through an anion exchange membrane (AEM) while Na^+ will move toward the cathode through a cation exchange membrane (CEM). This results in desalination of the saline water in the middle compartment.

The key difference between ED and distillation or RO is that ED removes salt ions from saline water while distillation and RO remove water from saline water. Considering the amounts of salt and water present in seawater, ideally ED should be able to achieve desalination with higher recovery of water at a lower cost than RO. However, while ED is the most cost-effective method for brackish water desalination, it is currently more expensive than RO for seawater desalination.[14] This is because the current electrode reactions used in ED cells to induce ion movement require a high operating voltage. ED performed in aqueous solutions typically utilizes water reduction to H_2 as the cathode reaction and Cl^- oxidation to Cl_2 or water oxidation to O_2 as the anode reaction.[14,21] The thermodynamic equilibrium cell voltages required to achieve water reduction/Cl^- oxidation and water reduction/water oxidation under standard conditions are 1.36 and 1.23 V, respectively. The actual operating voltages are significantly higher than these equilibrium cell voltages due to the kinetic overpotentials necessary to drive these reactions at desired rates in addition to other voltage losses (e.g., *IR* loss in solution and junction potentials across membranes).

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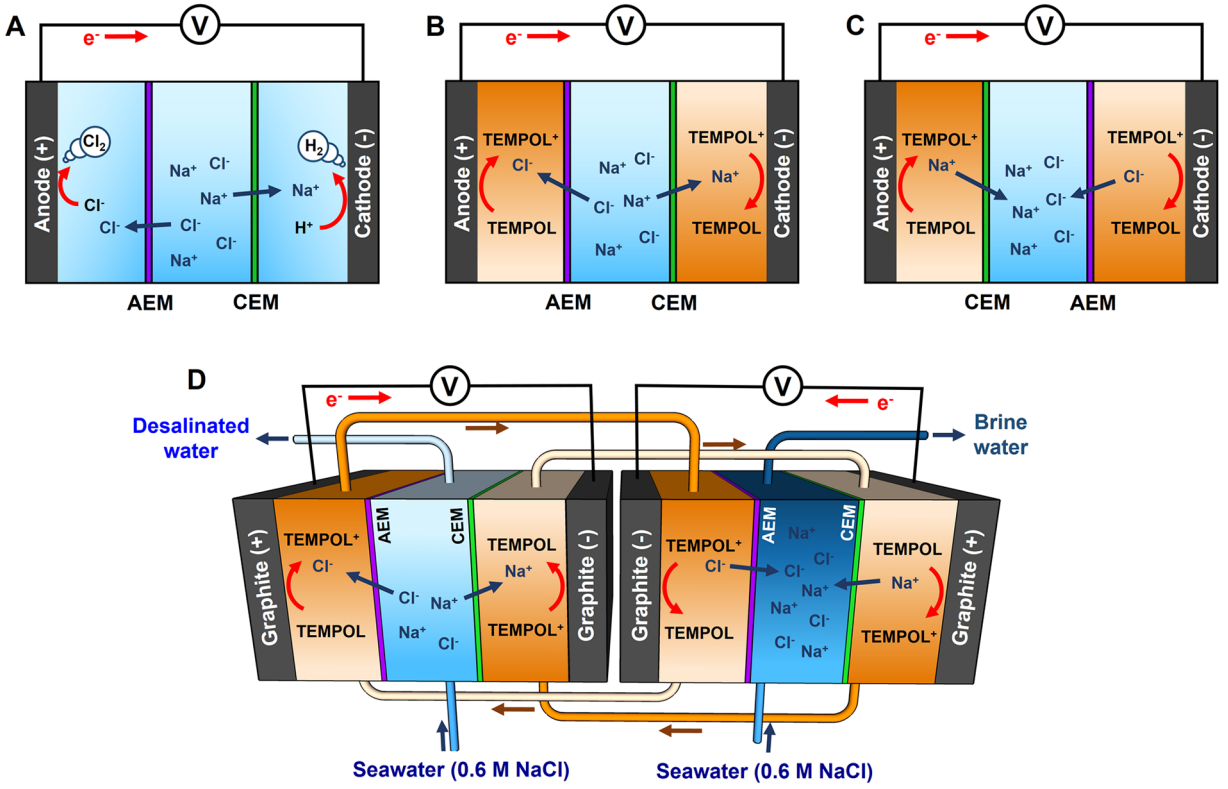

Figure 1. Schemes for (A) a conventional desalination ED cell, (B) a desalination ED cell based on the TEMPOL/TEMPOL$^+$ redox couple, (C) a salination ED cell based on the TEMPOL/TEMPOL$^+$ redox couple, and (D) combined desalination and salination ED cells in tandem to enable the sustainable use of the TEMPOL/TEMPOL$^+$ redox couple.

The electrical energy required to operate an ED cell is the product of the operating voltage and charge passed. Because the amount of charge that needs to be passed is determined by the salinity and volume of the feedwater, the only way to decrease the operating cost of ED to enable more cost-effective seawater desalination is to decrease the operating voltage of the cell by identifying new electrode reactions. In this study, we report a new type of ED cell where the oxidation and reduction of the same redox couple with fast redox kinetics are used as the anode and cathode reactions, respectively. This reduces the thermodynamic equilibrium cell voltage of the ED cell to 0 V while also minimizing the kinetic overpotentials. We demonstrate the sustainable use of the redox couple for ED by a new strategy that combines a desalination ED cell and a salination ED cell in tandem. The resulting tandem desalination/salination ED cell can successfully convert a 0.6 M NaCl solution (salinity of seawater) to fresh water in the desalination cell and 1.2 M NaCl in the salination cell without accumulating NaCl in the anolyte or catholyte which contain the redox couples. The operating principles, construction, and performance of a proof-of-concept tandem desalination/salination system reported in this study are expected to provide new possibilities for more cost-effective electrochemical seawater desalination along with various other electrochemical desalination approaches that have been reported recently.[5−10,23−28]

2. EXPERIMENTAL SECTION

Materials. NaCl (99%, Sigma Aldrich) and 4-hydroxy-2,2,6,6-tetramethylpiperidine 1-oxyl (TEMPOL) (97%, Sigma Aldrich) were used without further purification. Deionized water (Barnstead E-pure water purification system, resistivity >18 MΩ cm) was used to prepare all solutions.

Preparation of Electrolytes Containing TEMPOL/TEMPOL$^+$. In this study, a 0.6 M NaCl solution containing both 50 mM 4-hydroxy-2,2,6,6-tetramethylpiperidine 1-oxyl (TEMPOL) and 50 mM oxidized 4-hydroxy-2,2,6,6-tetramethylpiperidine 1-oxyl (TEMPOL$^+$) was commonly used as the anolyte and catholyte of the ED cells. To prepare this solution, the following electrochemical procedure was used. A three-electrode setup composed of Pt foil as the working electrode, a second Pt foil as the counter electrode, and a Ag/AgCl (4 M KCl) electrode as the reference electrode was used. The electrolyte in the working electrode compartment was 11 mL of a 0.6 M NaCl solution containing 0.1 M TEMPOL. The electrolyte in the counter electrode compartment was 11 mL of a 0.6 M NaCl solution. The working electrode compartment and the counter electrode compartment were divided by an anion exchange membrane (AEM) (PEEK, Fumatech). The complete electrochemical oxidation of TEMPOL to TEMPOL$^+$ was performed at room temperature with stirring by applying a potential of 1.0 V vs Ag/AgCl until the anodic current became 0 mA cm^{-2}, indicating the complete oxidation of TEMPOL to TEMPOL$^+$. Then, the resulting 0.6 M NaCl solution containing 0.1 M TEMPOL$^+$ was mixed with an equal volume of 0.6 M NaCl solution containing 0.1 M TEMPOL to form a 0.6 M NaCl solution containing 50 mM TEMPOL and 50 mM TEMPOL$^+$.

Construction and Operation of Desalination/Salination ED Cells. A custom-built Teflon cell was used for electrodialysis (ED) tests. The cell was composed of three compartments (anode compartment, middle compartment, and cathode compartment), and an AEM or CEM could be inserted as desired between any two compartments. For the desalination cell, an AEM (PEEK, Fumatech) was placed between the anode and middle compartments, and a CEM (Nafion 1110, Fumatech) was placed between the middle and cathode compartments. For the salination cell, a CEM (CMI-7000, Membranes International) was placed between the anode and middle

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compartments, and an AEM (PEEK, Fumatech) was placed between the middle and cathode compartments. The inner volume of the anode, middle, and cathode compartments was ~1 mL for each chamber. The area of the graphite anode and graphite cathode exposed to the electrolyte was 1 cm^2. The area of the AEM and CEM exposed to the electrolyte was also 1 cm^2.

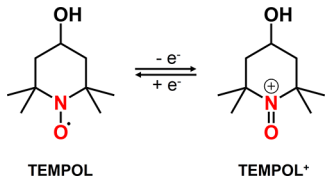
A 0.6 M NaCl solution containing 50 mM TEMPOL and 50 mM $TEMPOL^+$ was used as the anolyte and catholyte of both the desalination and salination ED cells. The anolyte of the desalination cell and the catholyte of the salination cell (combined volume of 4.8 mL) were circulated by using a peristaltic pump (BT100-2J, Longer pump) with a flow rate of 35 mL min^{-1}. The catholyte of the desalination cell and the anolyte of the salination cell (combined volume of 4.8 mL) were circulated in the same manner. The water to be desalinated in the middle compartment (0.6 M NaCl) of the desalination cell (volume of 3.58 mL) was circulated by itself by using the same peristaltic pump to enhance the mass transport of salt ions. The water to be salinated in the middle compartment (0.6 M NaCl) of the salination cell (volume of 3.58 mL) was also circulated by itself in the same manner. The salinity of the desalinated and salinated water was examined by using a salinity meter (Horiba D-74). Additionally, a chloride ion meter (Horiba 6560-10C) and a sodium ion meter (Horiba B-722) were used to confirm the results obtained with the salinity meter.

Operation of Desalination/Salination ED Cells with a Si Solar Cell. To demonstrate the operation of the desalination/salination ED cells powered solely by a solar cell that provides a limited voltage (<0.6 V), which is not sufficient to operate conventional ED cells, a NREL certified single junction Si cell (Newport) was used. The size of the Si solar cell was 2 cm × 2 cm (4 cm^2). The open-circuit voltage (V_{OC}) and short-circuit current density (J_{SC}) of the Si cell were 0.563 V and 35 mA cm^{-2}, respectively. The light used to illuminate the solar cell was generated from an Oriel LCS-100 solar simulator (100 W Xe arc lamp) equipped with an AM1.5G filter. The intensity of the light was calibrated to be 1 sun (100 mW cm^{-2}) at the surface of the Si solar cell, which was confirmed by a NREL certified GaAs reference cell (PV Measurement). Both the desalination ED cell and the salination ED cell were connected to the same Si solar cell in parallel as the electrical power generated by a single Si solar cell was sufficient to operate both of the cells.

3. RESULTS AND DISCUSSION

We selected the redox reactions of 4-hydroxy-2,2,6,6-tetramethylpiperidine 1-oxyl (TEMPOL) and its oxidized species ($TEMPOL^+$) (Scheme 1) as the electrode reactions to operate our ED cell (i.e., oxidation of TEMPOL at the anode and reduction of $TEMPOL^+$ at the cathode) (Figure 1B). By use of the forward and reverse of the same electrode reaction in solutions containing equal concentrations of TEMPOL and $TEMPOL^+$, the equilibrium potential of the anode reaction (E^e_{anode}) becomes equal to the equilibrium potential of the cathode reaction ($E^e_{cathode}$). As a result, the thermodynamic equilibrium potential of the ED cell, which is $E^e_{cathode} - E^e_{anode}$, is reduced to 0 V. In addition, the redox reactions of TEMPOL/$TEMPOL^+$ involve only a one electron transfer with fast kinetics (Scheme 1),[29,30] which minimizes the overpotentials required to drive the electrode reactions. By minimizing both the thermodynamic equilibrium potential and the kinetic overpotentials using the TEMPOL/$TEMPOL^+$ couple, we achieved the lowest possible operating voltage for the ED cell. TEMPOL and $TEMPOL^+$ are chemically stable in neutral aqueous solutions,[29,30] and TEMPOL is one of the cheapest nitroxyl radical derivatives.[31]

Scheme 1. Redox Reaction of TEMPOL and $TEMPOL^+$



Before the construction of an ED cell, the redox properties of a 0.6 M NaCl solution (mimicking the salinity of seawater) containing 50 mM TEMPOL and 50 mM $TEMPOL^+$ were first examined by cyclic voltammetry (CV). These CVs were obtained in an undivided cell (Figure 2A) by using graphite electrodes as both the anode and cathode and a Ag/AgCl reference electrode. From this CV, the half-wave potential ($E_{1/2}$) was estimated to be 0.60 V vs Ag/AgCl (4 M KCl), which was equivalent to the open-circuit potential of the solution (E_{OC}).

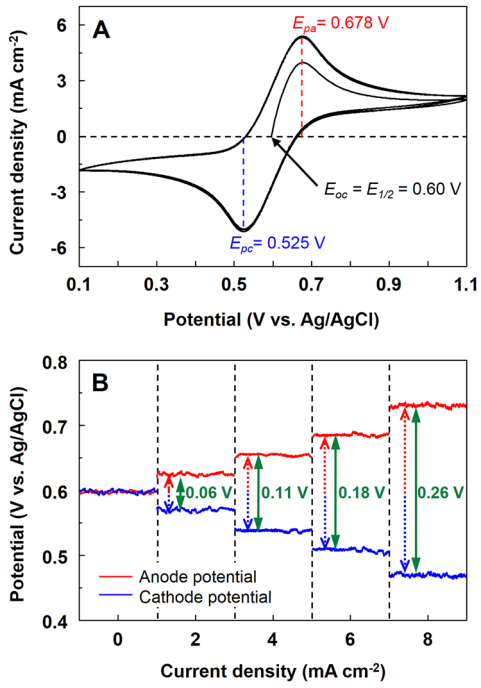


Figure 2. (A) CV obtained in a 0.6 M NaCl solution containing 50 mM TEMPOL and 50 mM $TEMPOL^+$ with a scan rate of 10 mV s^{-1}. (B) Biases between the graphite anode and graphite cathode required to generate current densities ranging from 0 to 8 mA cm^{-2} (indicated with green arrows). Individual anode and cathode potentials vs the Ag/AgCl reference electrode are also shown (dashed red and blue arrows, respectively).

Figure 2B shows the voltage applied between the graphite anode and graphite cathode to achieve various current densities. For example, to generate a current density of 8 mA cm^{-2}, a bias of only 260 mV was needed. Figure 2B also shows the individual potentials of the anode and cathode measured against the Ag/AgCl reference electrode at each bias condition. Because the kinetics for the oxidation of TEMPOL and the reduction of $TEMPOL^+$ are equally fast and the solution contains a 1:1 ratio of TEMPOL to $TEMPOL^+$, the cell voltage is symmetrically partitioned to the anode and cathode reactions with respect to the E_{OC} of the solution.

A desalination ED cell constructed based on the TEMPOL/$TEMPOL^+$ redox couple is shown in Figure 1B. It contains

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three compartments, where the anode compartment and cathode compartment sandwich a middle compartment containing a 0.6 M NaCl solution. The anolyte and catholyte were both composed of 0.6 M NaCl containing 50 mM TEMPOL and 50 mM $TEMPOL^+$. The anode compartment and middle compartment were divided by an AEM, while the cathode compartment and middle compartment were divided by a CEM. When a voltage was applied between the graphite anode and graphite cathode, the oxidation of TEMPOL to $TEMPOL^+$ occurred at the anode, causing Cl^- to move from the middle compartment to the anolyte. At the same time, reduction of $TEMPOL^+$ to TEMPOL occurred at the cathode, causing Na^+ from the middle compartment to move to the catholyte, thus achieving desalination of the middle compartment. We note that ion removal in ED is not ion-type specific because ion movement in ED occurs to maintain charge neutrality in all compartments, compensating for the changes in charge caused by the cathode and anode reactions. Thus, if the middle compartment contains ions other than Na^+ and Cl^-, like real seawater, they can also be removed.

The major problem with sustaining operation of the ED cell shown in Figure 1B is that although the anolyte and catholyte initially contain equimolar TEMPOL and $TEMPOL^+$, the solution composition changes once current starts to flow. The ratio of TEMPOL:$TEMPOL^+$ will gradually decrease in the anolyte but increase in the catholyte. Because of these concentration changes, the equilibrium voltage to operate the cell gradually increases above 0 V. Eventually, the anolyte contains only $TEMPOL^+$ and the catholyte contains only TEMPOL, causing the cell operation to terminate before desalination is complete. Simply swapping the anolyte and catholyte to reinitiate the cell operation does not work as a long-term solution because Cl^- and Na^+ are also accumulated in the anolyte and catholyte, respectively. Therefore, if the two solutions are simply swapped, both the anolyte and catholyte will eventually contain a high concentration of NaCl. The processes required to recover TEMPOL and $TEMPOL^+$ from the brine anolyte and catholyte would make such an ED cell impractical.

To resolve this issue, we constructed an additional ED cell that utilizes the same anode and cathode reactions but performs salination of the feedwater rather than desalination of the feedwater. This is achieved by switching the positions of the AEM and CEM as shown in Figure 1C. The strategy employed here is to operate the desalination and salination ED cells in tandem so that any composition changes that occur in the catholyte and anolyte of the desalination ED cell can be reversed in the salination ED cell (Figure 1D). More specifically, the anolyte of the desalination cell and the catholyte of the salination cell are circulated so that the change in TEMPOL/$TEMPOL^+$ concentration and Cl^- accumulation in the anolyte of the desalination cell are reversed in the catholyte of the salination cell. In the same manner, the catholyte of the desalination cell and the anolyte of the salination cell are circulated so that the change in TEMPOL/$TEMPOL^+$ concentration and Na^+ accumulation in the catholyte of the desalination cell are reversed in the anolyte of the salination cell. This allows for the ED operation to continue until all NaCl in the feedwater of the desalination cell is completely removed. Furthermore, Na^+ and Cl^- removed from the feedwater in the desalination cell do not accumulate in the anolyte and catholyte of the desalination cell. Instead, they are collected in the brine solution generated by the salination cell. This eliminates the need to recover TEMPOL and $TEMPOL^+$ from the brine anolyte and catholyte while effectively concentrating NaCl to a desired volume in the salination cell.

The tandem operation of desalination/salination ED cells was investigated while applying 0.6 V between the anode and cathode for both the desalination and salination cells. The feedwater for both the desalination and salination cells was 0.6 M NaCl. The current density−time (J−t) plots and the charge passed−time (Q−t) plots for both the desalination cell and salination cell are shown in Figure 3A. Initially, the J−t plots of

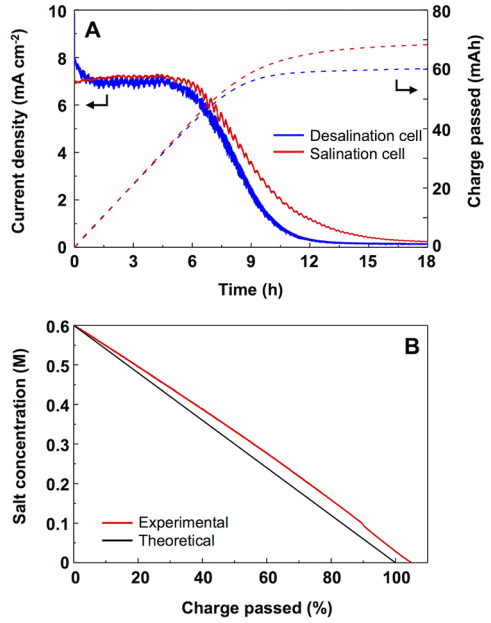

Figure 3. (A) J−t plot (solid) and corresponding Q−t plot (dashed) obtained for the desalination cell (blue) and salination cell (red) at an operating voltage of 0.6 V. (B) Change in salt concentration measured in the middle compartment of the desalination cell during ED operation.

the desalination and salination cells showed constant and comparable current density generation, confirming that our strategy to operate the desalination/salination cells in tandem was successful in maintaining equal TEMPOL/$TEMPOL^+$ compositions in the anolyte and catholyte.

As desalination progressed, the current densities of both the desalination and salination cells decreased gradually. The decrease in the current density of the desalination cell is due to the decrease in salt concentration in the middle compartment. As the salt concentration decreases, ion movement is retarded out of the diluted middle compartment, and as a result, the current density of the desalination cell decreased. The current density of the desalination cell eventually becomes zero when desalination of the feedwater is completed.

As the current density of the desalination cell decreased, the current density of the salination cell also decreased; however, the decrease in current density of the salination cell has a different origin. In the salination cell, the salt concentration of the middle compartment was increasing during operation, so the current should not have decreased as it did for the desalination cell. However, complete reversal of the changes in TEMPOL/$TEMPOL^+$ concentration generated in the salination cell could not be achieved in the desalination cell because

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of the decreased reaction rate in the desalination cell. As a result, the ratio of TEMPOL/TEMPOL$^+$ in both the anolyte and catholyte started to change, and the equilibrium cell voltage of the salination cell increased. Because the cell was being operated at a fixed potential, the overpotentials for both the cathode and anode reactions decreased, causing the current density to decrease. Eventually, the current density of the salination cell decayed to zero when the anolyte contained only TEMPOL$^+$ and the catholyte contained only TEMPOL. The extra charge passed in the salination cell shown in Figure 3A matched exactly with the charge required for the conversion of 1:1 TEMPOL/TEMPOL$^+$ to only TEMPOL in the catholyte and TEMPOL$^+$ in the anolyte.

The amount of charge passed during ED cell operation must be directly proportional to the amount of salt ions removed from the feedwater (i.e., removal of one Na^+ ion and one Cl^- ion per electron) in the desalination cell. This is because current cannot flow through the ED cell without coupled ion (Na^+ and Cl^-) movement out of the feedwater. This was confirmed by monitoring the salinity of the feedwater by using a salinity meter as a function of charge passed (Figure 3B). The salinity of the feedwater became zero when 105% of the stoichiometric amount of charge to remove all Na^+ and Cl^- from the feedwater was passed (Figure 4), demonstrating the

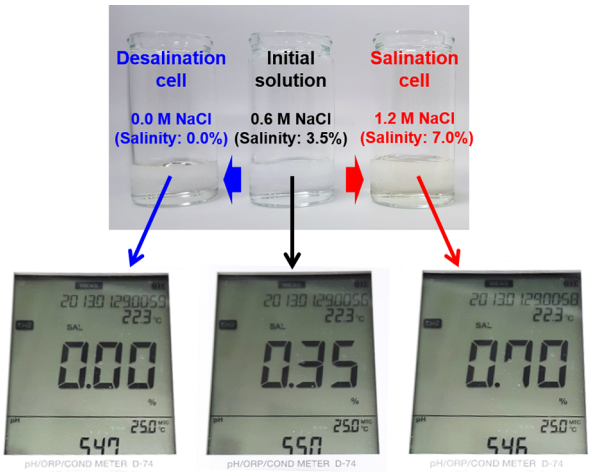

Figure 4. Photographs showing the salinity change of the saline water in the middle compartment before and after cell operation. The salinity of each solution was measured by using a salinity meter, which shows the salinity as wt % of NaCl in the solution. 0.6 and 1.2 M NaCl are equivalent to 3.5 and 7.0 wt % salinity, respectively. The salinity measurements were obtained after solutions were diluted by a factor of 10 so that the salinities of these solutions were within the measurement range of the salinity meter. We note that considering the resolution of the salinity meter (0.01%) and the dilution factor, the maximum NaCl concentration of the desalinated water can be 0.0086 M when the salinity meter shows 0.00% salinity.

expected behavior of the desalination ED cell. In the same manner, charge passed in the salination cell is coupled with the addition of salt ions into the feedwater. Indeed, the NaCl concentration of the feedwater in the salination cell was confirmed to be 1.2 M when the cell operation was completed (Figure 4).

The complete conversion of a 0.6 M NaCl solution to fresh water in the desalination cell and a 1.2 M NaCl solution in the salination cell, without accumulation of NaCl in the anolyte and catholyte containing the TEMPOL/TEMPOL$^+$ couple, confirmed that our tandem desalination/salination strategy can successfully enable the sustainable and practical use of redox couples for ED applications. Furthermore, we note that although the TEMPOL/TEMPOL$^+$-based ED cells can completely desalinate 0.6 M NaCl at 0.6 V, conventional ED cells cannot even operate at 0.6 V because their thermodynamic equilibrium cell voltages are already ≥1.23 V.

The significant reduction of the operating voltage of our TEMPOL/TEMPOL$^+$-based ED cells has two important implications. The first is that practical seawater desalination by ED can be enabled, as the high operating voltage of conventional ED cells is the major limiting factor that inhibits the use of ED for seawater desalination. The second is that due to the significant reduction of the operating voltage, our ED cell can operate with inexpensive portable power generators (e.g., batteries and solar cells) that can provide only a limited voltage. This can be particularly beneficial when desalination needs to be achieved in regions where grid-based electricity is not available. Because the thermodynamic equilibrium potential of our ED cells is 0 V, ideally, any power generator that can provide a voltage >0 V can be used to operate our ED cells. The results shown in Figure 3A indicate that with our current cell setup a battery or a solar cell that can offer ~0.6 V can operate the TEMPOL/TEMPOL$^+$-based ED cell with a current density level of 6−8 mA cm^{-2}. (The current density can be increased further when a high surface area electrode instead of a flat graphite electrode is used.)

For example, Figure 5A demonstrates the use of an inexpensive single junction Si cell to operate our TEMPOL/TEMPOL$^+$-based tandem ED cell solely by illuminating the solar cell using light mimicking solar illumination (AM1.5G, 100 mW cm^{-2}) with no external energy input. The $J-V$ characteristics of the single junction Si cell used in this study are shown in Figure 5B. The V_{OC} and J_{SC} were found to be 0.563 V and 35 mA cm^{-2}, respectively. Although the V_{OC} of a single junction Si solar cell is very limited, the electrical power generated by one Si solar cell is sufficient to operate both the desalination and salination ED cells connected in parallel to one Si solar cell. The $J-V$ and $J-Q$ plots obtained for the desalination/salination ED cells operated by the Si solar cell are shown in Figure 5C. They are comparable to those shown in Figure 3A with slightly lower current densities, which was expected because the operating cell voltage provided by the solar cell was lower than 0.6 V.

The successful conversion of a 0.6 M NaCl solution to 0 and 1.2 M NaCl solutions in the desalination cell and salination cell, respectively, by using only an inexpensive single junction Si cell with a limited V_{OC} clearly highlights the advantages offered by our new ED strategies. The new ED strategies demonstrated with a TEMPOL/TEMPOL$^+$ redox couple in this study may be extended to the use of various types of redox couples with fast kinetics. We tested crossover of TEMPOL and TEMPOL$^+$ through the AEM and CEM for an extended period of time (24−48 h) and found that they can cross through both the AEM and CEM. Thus, membranes that can completely block the crossover of TEMPOL and TEMPOL$^+$ must be developed in the future or redox couples that do not suffer from crossover problems may be selected for more practical ED operation. The rapid progress made in flow cell batteries for large-scale stationary energy storage,[29,32] which are also based on the use of various redox couples and compatible membranes, is expected to synergistically expedite the further development of appropriate membranes and the

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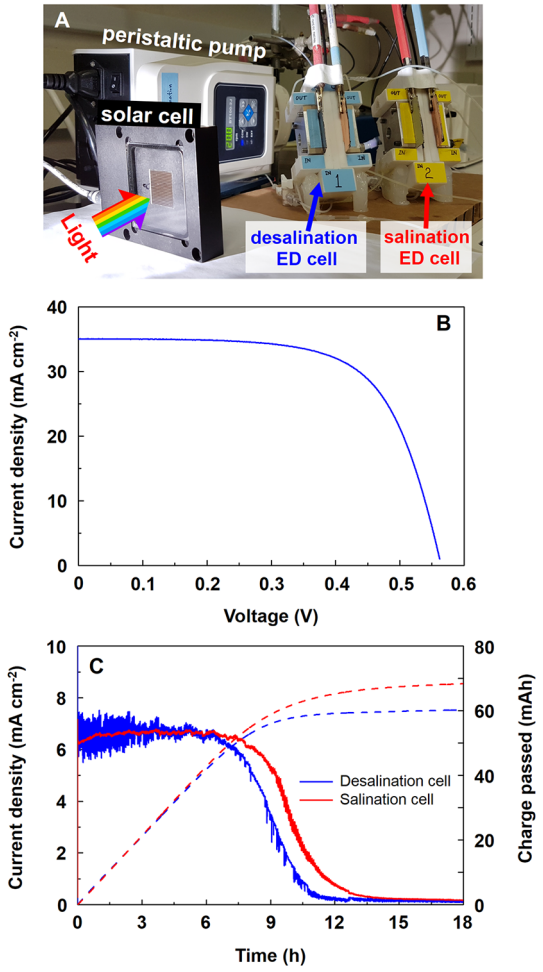

Figure 5. (A) Photograph showing the experimental setup for the tandem ED cells operated by a single junction Si solar cell. (B) $J-V$ characteristics of a single junction Si solar cell under AM1.5G, 100 mW cm^{-2} illumination. (C) $J-t$ plot (solid) and corresponding $Q-t$ plot (dashed) obtained for the desalination (blue) and salination (red) ED cells powered solely by a single junction Si solar cell under AM1.5G, 100 mW cm^{-2} illumination.

proof-of-concept tandem ED system demonstrated in this study.

4. CONCLUSION

In summary, we demonstrated new ED strategies where the use of the TEMPOL/TEMPOL$^+$ redox couple and the operation of a desalination cell and a salination cell in tandem enabled sustainable desalination with a significantly decreased operating voltage compared to traditional ED cells. These strategies reduced the thermodynamic equilibrium cell potential to 0 V while also significantly reducing kinetic overpotentials required for cell operation. As a result, the overall operating voltage of our ED cell is remarkably reduced, making it possible to use ED for seawater desalination and also to operate ED cells using inexpensive portable power generators (e.g., batteries and solar cells) that provide a limited voltage. The tandem operation of desalination and salination cells enabled conversion of the feedwater (0.6 M NaCl) to 0 and 1.2 M NaCl solutions in the desalination cell and salination cell, respectively. This was achieved without the accumulation of salt ions in the anode and cathode compartments, which is critical for the sustainable use of the TEMPOL/TEMPOL$^+$ redox couple in the ED cells. The operating principles and performance of a proof-of-concept tandem desalination/salination system demonstrated in this study can lead to a multitude of new opportunities for the development of practical electrochemical desalination devices for seawater desalination as well as brackish water desalination.

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Notes

The authors declare no competing financial interest.

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