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**FUTURE ENERGY** 

#### Water Desalination with Energy Storage Electrode Materials

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As population demands for freshwater increase, existing natural freshwater resources face significant strains. Currently, over 2.5 billion people live in localities that are subject to severe water scarcity at least 1 month of the year.<sup>1</sup> Scarcity affects all types of localities, such as urban, rural, coastal areas, landlocked areas, and off-grid locations. Increasingly, active water purification technologies are being used to boost and secure freshwater supplies. A widely used desalination technology is seawater reverse osmosis (SWRO), in which pumps pressurize the feedwater to well above its osmotic pressure to pump water molecules through a membrane largely impermeable to salt ions ( $p_{osmotic} \sim 25$  bar).<sup>2</sup> City-scale SWRO plants are operational in several countries, delivering on the order of 10<sup>6</sup> m<sup>3</sup> of treated water per day (<0.1% of the total global daily water consumption). However, as the need for water purification increases and the requirements for each locality becomes more diverse, SWRO plants alone cannot meet the growing demand for a technological solution. Barriers toward increased penetration of SWRO include the enormous investment required to develop such plants, poor downscalability of the technology, the geographical limitation to coastal areas and near urban environments, and high energy requirements (typically about 4 kWh/m<sup>3</sup>).<sup>2</sup>

Electrochemical systems are mainly associated with energy storage, with well-known examples including batteries and supercapacitors. However, other electrochemical systems, such as electrodialysis (ED) and capacitive deionization (CDI), have long been identified as promising solutions for energy- and infrastructure-efficient brackish water desalination ( $\sim$ 0.5-30 g/L total dissolved solids [TDS]). Such systems desalt feedwaters by applying electric fields to the feed channel, and so removing salt ions by processes such as electromigration and electrosorption. Thus, electrochemical systems can desalinate at sub-osmotic pressures, and as a result can be highly scalable with no highpressure pumps required. ED relies on often-expensive ion-exchange membranes to desalt feedstreams and is today considered a mature technology.<sup>3</sup> By contrast, CDI has traditionally relied on inexpensive nanoporous carbon electrodes to desalt (such as activated carbons), and the CDI research field is rapidly growing and evolving.<sup>4</sup> A conventional CDI cell consists of two nanoporous carbon electrodes with a separator between them,



**Figure 1. Concept and Performance Values of CDI Cells with Capacitive and Faradaic Electrodes** A historical evolution of the salt storage capacity of desalination cells achieved with either nanoporous carbon electrodes (black circles), Faradaic electrodes (blue triangles), or hybrid cells with one nanoporous carbon electrode and one Faradaic electrode (red diamonds), with all values reported per mass of both electrodes.<sup>4–20</sup> Despite their recent introduction, cells with Faradaic electrodes have achieved significantly higher gravimetric salt storage capacities compared with cells with nanoporous carbon electrodes. Schematics contrast the salt storage mechanism of CDI cells leveraging nanoporous carbon (capacitive) electrodes and cells with Faradaic electrodes, such as layered intercalation electrodes or conversion electrodes.

where the separator also serves as the feed flow channel. During CDI cell charging, a voltage of around 1 V is applied to the cell, resulting in salt ions being removed from the feed and stored capacitively within electric double layers (EDLs) in the nanopores of the oppositely charged electrode (Figure 1). During charging, along with storage of oppositely charged ions (counterions), salt ions with similar charge to the electrode (co-ions) are expelled from the nanopores and into the feedwater. Co-ion expulsion is a parasitic loss, resulting in the ratio of moles of salt removed from the feed to moles of charge stored by the cell to be lower than unity, and so this effect must be minimized in CDI.<sup>4</sup> Once the cell is fully

charged, it must be discharged to allow ions to be released from the EDLs and enter the feed, leading to a brine cell effluent.

CDI cells share many similarities with aqueous electric double-layer capacitors (EDLCs), a type of supercapacitor that is commonly used for energy storage. Both technologies use nanoporous carbon electrode materials, operate within a limited voltage window bounded by the voltage for water splitting (ca. 1.2 V cell voltage; dependent on local pH and electrode material), store ions in nanopore EDLs, and undergo charge/discharge cycling.<sup>4</sup> Furthermore, due to their similarity with aqueous EDLCs, CDI cells store energy while desalinating and release it during electrode regeneration (discharging). Over the past decade, the field of CDI has been buoyed by the parallel development of nanoporous carbons for supercapacitors,<sup>21</sup> which has contributed to the order of magnitude improvement in CDI cell desalination performance and energy efficiency over that time period.<sup>4</sup> However, the performance of CDI systems employing nanoporous carbon electrodes has in the past couple of years largely plateaued (Figure 1), and the challenges facing this technology are becoming clearer. While CDI is a highly efficient technology for desalting feeds on the dilute end of the brackish water regime  $(\sim 0.6-3 \text{ g/L TDS})$ , it struggles with feedwater of higher salinity due to a limited capacity for ion storage and excessive co-ion expulsion.<sup>4</sup> Another challenge is a limited cycle life due to corrosion of the positively charged carbon electrode during cell charging, especially when cells are operated at voltages well above 1 V. CDI cells typically achieve less than 1,000 charge/ discharge cycles,<sup>22</sup> with cycle life depending on the type and composition of the feedwater stream, the electrode material, cell design, and other operational parameters.<sup>23</sup>

To develop next-generation electrochemical systems for water desalination, it is convenient to take inspiration from the highly developed energy storage field. As demonstrated by CDI cells, energy storage electrodes can be successfully applied as efficient water desalination electrodes (while maintaining their energy storage functionality). A large and promising category for exploration are the materials that store ions via processes other than capacitively in charging EDLs, which we will call Faradaic electrodes. An example of such a process is ion intercalation, the reversible insertion of cations or anions into sites within the solid electrode material. batteries and supercapacitors, In

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ion intercalation materials are often used as high-capacity electrodes. The most prominent examples are the electrodes used in commercial lithium-ion batteries, including lithium-ion intercalating graphite anodes, and cathodes such as lithium manganese oxide. Other examples include the intercalation cathodes used in sodium ion batteries, such as sodium manganese oxide (NMO).<sup>24</sup> To understand why Faradaic electrodes are highly attractive for water desalination, it is important to contrast how ions are stored in such electrodes to storage in nanoporous carbons (Figure 1). While in nanoporous carbons, salt ions are stored capacitively in EDLs along surfaces or in nanopore volumes, in intercalation electrodes, ions are stored within the electrode material in crystallographic sites or between atomic planes. This brings two important features. First, accessing storage within the electrode material can enable higher salt storage capacity than achieved with storage in EDLs, analogous to the higher energy density achieved by Faradaic electrodes compared with EDLC electrodes in energy storage systems. Second, unlike nanoporous carbon electrodes, Faradaic electrodes typically do not suffer from significant co-ion expulsion and so may allow for effective desalination even at seawater levels of feed salinity.

Faradaic electrodes have only recently begun to be explored toward water desalination. In pioneering work in 2012, Pasta et al.<sup>5</sup> presented the concept of a "desalination battery" using two Faradaic electrodes for feedstream desalination, including sodium-ion intercalation into a charging NMO cathode and chloride ion removal via oxidation of a silver metal electrode (a reversible conversion reaction, Ag/AgCl). This cell was highly innovative but impractical, as an electrode consisting of silver metal is cost prohibitive for large-scale applications. The concept was further developed in 2014, by Lee et al.,<sup>6</sup> who replaced the silver electrode with a nanoporous carbon and anion exchange membrane assembly, introducing the concept of "hybrid CDI" as a cell that uses one EDLC electrode and one Faradaic electrode. The authors demonstrated that such a cell can store up to 31.2 mg of salt per gram of electrode material during cell charging, about 50% higher than the salt storage achieved by classical CDI cells using solely nanoporous carbon electrodes (Figure 1). Subsequent hybrid CDI systems have paired positively charged nanoporous carbon electrodes with Faradaic cathodes such as sodium iron phosphate or two-dimensional layered titanium disulfide.<sup>7,20</sup> In 2016, Srimuk et al.<sup>8</sup> developed a cell with two-dimensional MXene intercalation electrodes as anode and cathode and demonstrated the first desalination cell relying entirely on the mechanism of ion intercalation. MXene is a material consisting of layered 2D transition metal carbide planes, with ions inserting between planes during charging. As such, they are promising for desalination applications as they can store other types of ions beyond solely sodium and possess high electric conductivity. MXene demonstrated significantly less capacity for anion storage compared with cation storage, and the measured overall cell salt storage capacity obtained was  $\sim$ 13 mg/g when using a 5 mM NaCl feed and charging to 1.2 V.<sup>8</sup> Another two-dimensional intercalating nanomaterial, now consisting of MoS<sub>2</sub> layers, was applied to water desalination by both Xing et al. and Srimuk et al. in 2017.<sup>9,10</sup> The stable use of  $MoS_2$  in a symmetric cell is limited to a charging voltage of 0.8 V due to the catalytic nature of MoS<sub>2</sub> in evolving hydrogen, and the cell of Srimuk et al. demonstrated a salt storage capacity of ~10 mg/g for 5 mM NaCl feedwater.

A novel direction for improved anion storage was proposed by both Nam et al.<sup>25</sup> and Chen et al.<sup>26</sup> in 2017, which was to use a bismuth conversion electrode to store chloride ions via oxidation to the insoluble metal oxychloride BiOCl. As shown through equilibrium calculations by Nam et al.,<sup>25</sup> BiOCl is stable and insoluble over a wide potential window for a pH range of 0-10.5 and inhibits parasitic water oxidation during charging. The calculated capacity of the electrode tested experimentally by Nam et al.,<sup>25</sup> in grams of chloride ions stored per gram of bismuth, was over four times the capacity for chloride ion storage calculated per gram of carbon electrode in a typical CDI cell. In a separate direction toward mitigating the issue of anion storage, several groups have recently developed desalination cells using sodium intercalation materials for both electrodes and separating the electrodes with an anion exchange membrane.<sup>11–13</sup> Relying just on cation intercalation, such a cell avoids anion intercalation altogether and had been previously postulated and described theoretically by Smith and Dmello.<sup>27</sup> In the flow compartment adjacent to the intercalating electrode, sodium ions were removed from the feed by the electrode and chloride ions electromigrated through the membrane, resulting in desalination of this flow. The opposite compartment accepted sodium ions from the de-intercalating electrode and chloride ions passing through the membrane, thus forming a brine solution. Proof-of-concept cells leveraging symmetric Prussian blue analogs for sodium intercalation, such as nickel hexacyanoferrate (HCF) or copper HCF, achieved a salt storage capacity of about 35 mg/g (for cells without membrane stacking),<sup>11,12</sup> and nearly 60 mg/g for an asymmetric system with one nickel HCF and one iron HCF electrode when calculated over the entire charge-discharge cycle.<sup>13</sup> The latter value for salt storage capacity is roughly three times higher than that achieved by CDI cells with nanoporous carbon electrodes.

While only about a dozen Faradaic materials have been tested as electrodes in

desalination cells, and many of these materials were proposed only in the past calendar year, these electrodes have regularly achieved gravimetric salt storage capacities far exceeding that of nanoporous carbon electrodes (Figure 1). In addition to improved storage, another feature of desalination cells using Faradaic electrodes is the low energy requirements of the first proof-of-concept cells. For example, Kim et al.<sup>12</sup> demonstrated an energy requirement of roughly 0.02 kWh per 1 m<sup>3</sup> of treated water for the desalination of 25 mM NaCl feedwater, roughly an order of magnitude lower than the reported energy requirements of a cell with nanoporous carbon electrodes for the same feed and effluent. The basis for the attainment of such low energy requirements may be that Faradaic electrodes can achieve exceptionally high salt adsorption capacities while operated at lower cell voltages compared with CDI cells, often well under 1 V.<sup>8,12,27</sup> Initial results are also promising toward desalting high salinity streams, such as seawater, a regime where CDI with nanoporous carbons is largely ineffective. For example, with a "rocking chair"-type cell, Lee et al.<sup>13</sup> demonstrated removal of 40% of ions from a small (0.6 mL) 500 mM NaCl feed volume consuming energy of 0.34 kWh/m<sup>3</sup>. Using layered MoS<sub>2</sub> electrodes, Srimuk et al.<sup>9</sup> showed that the per ion energy requirement for a 500 mM feed was  ${\sim}24$  kT per ion removed, in stark contrast to the energy required to desalinate the same feed with standard nanoporous carbon electrodes, which was >10,000 kT per ion. The latter value is tremendously large because nanoporous carbons cannot effectively desalinate high salinity streams due to the parasitic effect of co-ion current.<sup>4</sup> However, first results with intercalation electrodes suggest that these cells do not suffer from excessive co-ion currents even for very high salinity feeds.

The Faradaic electrodes investigated thus far toward desalination can be

broadly classified into materials that are highly tuned toward a specific ion, to the exclusion of other similarly charged ions, and materials that can potentially simultaneously store a multitude of ion types. Examples of ion-specific materials are those performing conversion reactions, such as conversion of Bi to BiOCl, or those with properly tuned tunnel structures. For example, Kim et al.<sup>28</sup> showed that their NMO electrode selectively removed Na<sup>+</sup> at a factor of 13 over K<sup>+</sup> when treating waters containing both these ions at equal initial concentration. Developing highly selective desalination systems, which target specifically key pollutants or scalants in feedwaters, such as Ca<sup>2+</sup>, is an important research direction in the larger desalination field.<sup>29</sup> Projecting into the future, novel selective electrodes can potentially be developed based on the needs of the water treatment community, for example, selectively storing water pollutants such as lead ions or boron. Less-specific electrodes have the potential to store a multitude of ions, and some even allowing cation and anion intercalation. For example, 2D layered MXene electrodes have been shown to remove effectively a wide variety of cations such as Na<sup>+</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, and Ca<sup>2+</sup>.<sup>30</sup> Materials that are not tuned to a single ion may be more suitable for desalination of complex feedstreams containing many ion types requiring removal, such as most brackish and seawater sources. Although the effective removal of several cations simultaneously has yet to be demonstrated with MXenes, this material is promising toward desalination of complex feedwaters.

One issue with Faradaic electrodes may be slower salt removal relative to nanoporous carbons, in close analogy to the energy storage field where intercalation electrodes typically exhibit lower power density relative to nanoporous carbon electrodes. Initial studies with desalinating intercalation electrodes are often limited to low (order

100  $\mu$ A/cm<sup>2</sup>) applied currents,<sup>11,13</sup> in contrast to CDI cells with nanoporous carbons, which typically desalt with applied currents greater than  $1 \text{ mA/cm}^{2.4}$  In the future, a careful comparison of salt removal kinetics is required between cells with Faradaic electrodes and classical CDI cells under similar experimental conditions, such as feedwater and effluent salt concentration, to draw rigorous conclusions. Many strategies developed in the energy storage field to improve the power density of intercalation electrodes will be pertinent for desalination electrodes. For example, the development of 3D materials with open foam structure or with small, accessible particles of intercalation materials will help maximize rate capability.<sup>25</sup> We must stress that we do not expect Faradaic electrodes to supplant nanoporous carbons in desalination systems. Just as Faradaic electrodes and EDLCs exist side by side as important energy storage technologies, also for desalination both electrode classes have unique advantages and disadvantages. For example, nanoporous carbons may remain the best option for applications where fast desalination is required, while intercalation materials will be required for desalination of higher salinity streams. Both nanoporous carbons and intercalation electrodes can be highly selective, but both rely on different selectivity mechanisms and thus may have relative advantages and disadvantages toward selectively removing key species from feedwaters. Nanoporous carbons, especially commercial activated carbons, can be very inexpensive, and thus classical CDI systems may remain highly attractive when considering desalination performance metrics normalized by electrode cost.

What is perhaps most exciting is that the exploration of Faradaic materials as desalination electrodes has just begun, largely in the past calendar year (Figure 1). Over the past decades, the energy storage field has developed

hundreds of potential candidate materials, a deep and promising well to explore for desalination.<sup>24,31–35</sup> Thus, much foundational work has already been accomplished by the energy community, as many promising Faradaic electrode materials have been synthesized and characterized for storage capacity, cycle life, and round-trip energy efficiency, albeit in battery or supercapacitor electrolytes. This includes vast research activities related to sodium and potassium batteries, and also work on anion intercalation materials.<sup>24,34,35</sup> Many possibilities for future advances exist, for example, future directions in chloride ion storage in desalination systems can take inspiration from the burgeoning field of chloride ion batteries. In such batteries, metal oxychlorides, including VOCI and FeOCI, have been investigated as cathode materials but have yet to be checked for their compatibility and performance in desalination cells.<sup>33</sup> On the other hand, much work remains toward the characterization and performance stability testing of already proposed materials. Many metrics important to desalination, such as cycle life in synthetic and real feedwaters, performance under conditions of biofouling, and obtainable water recovery, remain at present unknown for desalination systems based on Faradaic electrode materials. The rapid ascension of the field of CDI with Faradaic electrodes over the past calendar year is evidence that numerous research groups developing electrode materials for energy storage are becoming aware of their potential application in water treatment. We believe this field is on the threshold of achieving explosive growth and that energy storage electrode materials may very well be the source of leading, next-generation water treatment systems.

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