Voltage- and Flow-Controlled Electrodialysis Batch Operation: Flexible and Optimized Brackish Water Desalination

Wei He^{a,b,1,*}, Anne-Claire Le Henaff^{a,1}, Susan Amrose^a, Tonio Buonassisi^a, Ian Marius Peters^a, Amos G. Winter, V^a

^aDepartment of Mechanical Engineering, Massachusetts Institute of Technology, Cambridge, MA, 02139, United States ^bSchool of Engineering, University of Warwick, Coventry, CV4 7AL, United Kingdom

Abstract

Electrodialysis (ED) desalination has been demonstrated to be more energy-efficient, provide higher-recovery, and be lower-cost for producing drinking water from saline groundwater compared to reverse osmosis. These benefits of ED could translate into cost-effective, renewable-powered desalination solutions. However, the challenge of using a variable power source (e.g. solar) with traditional steady-state ED operation requires batteries to reshape the power source to match the desalination load; these batteries often contribute to a large fraction of the produced water cost. In this study, we propose a time-variant voltage- and flow-controlled ED operation that can enable highly flexible desalination from variable power sources, including renewables, with negligible batteries, potentially leading to reduced water costs compared to what existing technology can provide. A model-based controller is presented which varies applied ED stack voltage and pumping flow rate to match power consumption to a variable source while maximizing desalination rate throughout an ED batch. The utility of the controller was demonstrated with a pilot-scale system tested with brackish groundwater, which operated as expected under varying fixed power levels and a real solar irradiance profile. The pilot system achieved a production rate up to 45% higher than that of an equivalently sized traditional steady-state ED system.

Keywords:

Brackish Water Desalination, Variable Power, Electrodialysis, Variable Voltage and Flow, Model-Based Control

1. Introduction

Almost two-thirds of the world's population, approximately four billion people, face severe water scarcity during at least one month of the year [1]. Pressures from 5 population growth and climate change are expected to 6 exacerbate this water stress by increasing water demand as water supplies become more erratic and uncertain 8 [2]. One approach to mitigate water stress is to make 9 use of brackish groundwater, or groundwater with a to-10 tal dissolved solids (TDS) concentration above the taste 11 threshold (>500 mg/L). Brackish groundwater is preva-12 lent throughout the world [3, 4, 5] and is increasingly 13 being used in the Middle East and North Africa to meet 14 municipal water demand [6]. However, its use is limited 15

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by the high-cost of desalination, difficulties of managing large volumes of waste brine, and the high costs of integrating with off-grid energy sources [7]. These issues are most challenging in remote, off-grid, rural communities that are prevalent in countries such as India [8], 20 where the majority of those facing severe water scarcity 21 live [1].

Currently, the dominant method of desalinating brack-23 ish groundwater is reverse osmosis (RO) [9]. Wright et 24 al. demonstrated that photovoltaic (PV)-powered elec-25 trodialysis (ED) can be an energy- and cost-effective 26 alternative solution to RO for village-scale applications, 27 particularly suited to rural India [8]. ED has a lower 28 energy consumption per unit water produced compared 29 to RO (75% less at 1,000 mg/L and 30% less at 3,000 30 mg/L), and a greater water recovery ratio (nearly double 31 that of current village-scale RO systems) [8]. The high 32 energy efficiency of ED reduces its carbon footprint and 33 translates into a smaller, less expensive renewable power 34 systems than those required for off-grid RO, which could 35 reduce total water costs. The high water recovery could 36

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^{1.1.} Background 2

^{*}Corresponding a author

Email address: whe@mit.edu, Wei.He.2@warwick.ac.uk (Wei He)

also lower water wastage and brine management costs

³⁸ relative to RO. These combined features make ED a

³⁹ promising technology for cost-constrained communities

⁴⁰ in developing countries and water scarce regions [10].

Although ED is amenable to renewable power due to its low specific energy consumption for brackish water 42 desalination, remaining challenges arise from balancing 43 variable renewable power sources and electrical demand 44 for producing water. A traditional static ED system typ-45 ically operates at a constant voltage and flow rate, which 46 creates an inflexible electrical load that often requires 47 large battery banks to reshape the variable power input 48 (from a source such as solar) for meeting the desalination 49 demand throughout the day. As a result, batteries con-50 tribute a large fraction of the lifetime cost and the total 51 water cost for an off-grid ED system [11, 12, 13]. Similar 52 challenges are also faced ED systems powered by elec-53 trical grids with incorporated wind and solar sources; 54 to constantly meet electrical demands (including desali-55 nation), high-cost energy storage is essential to provide 56 the flexibility that cancels out the intermittence of the 57

⁵⁸ renewables [14, 15].

⁵⁹ 1.2. Benefits of time-variant desalination

To mitigate some of these challenges and costs asso-60 ciated with energy storage, this study proposes a time-61 variant ED operation by varying voltage and flow rate, 62 to catalyse the flexible use of variable power sources 63 with negligible batteries. Using the proposed flexible operation, these time-variant ED systems could produce 65 more water than demanded when excess power is avail-66 able (say on sunny time) and store it for periods when 67 power is not available (say on cloudy time); this ap-68 proach would effectively store energy as treated water, 69 rather than storing it in batteries [13]. The flexibility 70 in utilizing variable power for water production could 71 reduce battery capacity compared to that required by 72 73 traditional renewable energy-powered ED systems that have similar daily production rates, thereby potentially 74 reducing total water costs. 75

Flexible desalination operation also offers several ben-76 efits to on-grid desalination. It could enable the exploita-77 tion of variable electricity tariffs (particularly low tariffs 78 during off-peak times) to reduce energy costs for desalination, as off-peak electricity tariffs are often less 80 expensive than peak electricity tariffs [16]. Flexible 81 desalination can also aid in lowering costs and carbon 82 83 emissions of the electrical grid, as the flexible ED operation could help smooth intermittent renewable power 84 and lead to less energy storage required for the supply-85 demand balance. 86

1.3. Review of prior work

Several flexible operation strategies for desalination 88 have previously been explored for minimizing required 89 energy storage. Richards et al. [17] presented a flexible 90 RO brackish water desalination system under a charac-91 terized domain of operational variables, in which the sys-92 tem could directly utilize wind or solar power sources to 93 continuously produce water without batteries. This flex-94 ible RO system was later experimentally demonstrated to 95 produce water under wind power at various speeds [18] 96 and solar power at several irradiance levels [19, 20]. 97 Cirez et al. [21] developed a flexible PV-ED system 98 using an optimized PV module design, which was com-99 posed of multiple connected PV cells in series/parallel 100 that could vary voltage applied to the ED stack and max-101 imize energy transfer given available solar irradiance. 102 Malek et al. [22] demonstrated robust and stable de-103 salination performance in a lab-scale, directly-coupled, 104 wind-powered ED system under various wind speeds, 105 turbulence intensities, and periods of oscillation. Veza 106 et al. [23, 24] actively controlled the flow rate and 107 voltage of a wind-powered ED system by developing a 108 database of correlations between available energy, prod-109 uct concentration, flow rate, and voltage applied to the 110 two ED stacks in the system. 111

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Each of these systems adjusted water production rates 112 to maximize utilization of the variable power resource. 113 However, none of these prior studies presents a deter-114 ministic model for how to control the voltage and flow 115 rate of an ED system to utilize all available power from a 116 variable power source while maximizing water produc-117 tion rate. In water-stressed, cost-constrained settings, 118 the utility of maximizing production is to increase the 119 number of people who can gain access to potable wa-120 ter. Furthermore, maximizing water production from 121 a finite system size can lead to smaller, lower-cost ED 122 systems by improving the productivity per unit material 123 (e.g. membranes). 124

1.4. Objectives of this study

The objective of the present study is to develop and 126 demonstrate a highly-flexible, time-variant ED operation 127 strategy that can accommodate variable power sources. 128 This work is built on our previous research focused on 129 PV-ED system cost optimization with a flexible on-off 130 control strategy proposed by Bian et al.[13], which max-131 imizes utilization of solar power on a day-by-day basis 132 to reduce required battery capacity, and a voltage con-133 trolled strategy proposed by Shah et al. [25], which max-134 imizes water production rate by continuously changing 135 the voltage applied to an ED stack to operate near lim-136 iting current density throughout a batch. In the present 137

study, by introducing a new degree of freedom in control 138 flow rate - the proposed flexible ED operation can si-139 multaneously maximize drinking water production and 140 variable power utilization. This is achieved by actively 141 optimizing and controlling the voltage applied to an ED 142 stack and the flow rate through it. To create, validate, 143 and explore this highly-flexible ED technology, this pa-144 per we: 145

- codify the flexibility of batch ED operation given voltage- and flow-controlled operation, and their impact on water production;
- develop a model-based controller that simultane ously co-maximizes water production rate and vari able power utilization; and
- validate the controller using a pilot-scale timevariant ED system and benchmark its performance
 validate to computing a train ED supervision
- relative to conventional static ED operation.

Electrodialysis desalination and time-variant op eration

157 2.1. Electrodialysis desalination

ED is an electrochemical process that removes ions us-158 ing an external electric field with selective ion-exchange 159 membranes. In an ED system (Fig. 1), saline water 160 flows through an ED stack which contains a series of al-161 ternating anion exchange membranes (AEM) and cation 162 exchange membranes (CEM). AEMs only allow passage 163 of anions and CEMs only pass cations. With an elec-164 tric field applied over the ED stack, anions flow towards 165 the anode and cations towards the cathode. Therefore, 166 the placement of AEMs and CEMs in series selectively 167 controls the ion removal across the membranes, and pro-168 duces alternating channels of diluate and concentrate. 169

There are two types of ED operation: continuous, in 170 which a saline feed is desalinated within a single pass 171 through multiple ED stages (Fig. 1a); and batch, in 172 which diluate and concentrate are recirculated through a 173 single-stage ED stack until the diluate is desalinated to 174 a desired product concentration (Fig. 1b). Compared to 175 continuous ED, batch ED potentially requires a smaller 176 stack, a smaller footprint, less membrane area, and lower 177 capital costs to build a small-scale desalination system 178 [13, 26, 25]. Based on these advantages, this study 179 focuses on batch ED operation. 180

2.2. The concept and advantages of voltage- and flow controlled ED operation

Figure 2 illustrates the advantages of voltage- and flow-controlled ED operation, namely through improved



Figure 1: Schematic of ED desalination operation. In ED, an electric field is applied across alternating cation (CEM) and anion (AEM) exchange membranes to transport ions from the diluate channels to the concentrate channels. In a continuous ED system (a), feed is often passed through multiple ED stacks to produce product water. In an batch ED system (b), diluate and concentrate are recirculated through a single ED stack until the diluate is desalinated to a desired product concentration.

operational flexibility and water production compared to 185 conventional static ED operation and voltage-controlled 186 ED operation. The term "flexibility" in this study refers 187 to the variability of power at which the ED system is 188 able to operate. Figure 2a depicts a typical power con-189 sumption pattern during a static ED batch, in which a 190 constant voltage and a constant flow rate are applied. 191 At each diluate concentration, as the batch desalinates 192 from feed to product, the power consumption is fixed 193 regardless of input power available. Static ED operation 194 does not have any flexibility, requiring the power source 195 (e.g. the grid or a solar system with batteries) to be able 196 to match the fixed desalination power demand. Figure 197 2d depicts the current density throughout a batch static 198 ED process, which represents the ion transfer rate across 199 the membranes. With higher applied current density 200 the system can desalinate faster. The limiting current 201 density determines the maximum salt removal rate be-202 fore splitting water [25]. To avoid water splitting, in a 203 conventional static ED batch, the applied voltage is de-204 termined by setting the current density below the most 205 constraining limiting current density, which occurs at 206 the end of the batch (Fig. 2d). As a result of this con-207 straint, the applied current density is much lower than 208 limiting at other points in the batch process, resulting 209 in underutilized capacity of the membranes throughout 210 much of the batch, and a salt removal rate that is lower 211



Figure 2: The operational domains of static, voltage-controlled, and voltage- and flow-controlled ED batch operations. (a) The power curve of a conventional static ED batch process (solid line). (b) The flexible power domain of a voltage-controlled ED batch process with constant flow rate (shaded area). (c) The flexible power domain of a voltage- and flow-controlled ED batch process (shaded area). (d) Applied current density and limiting current density of a conventional static ED batch process. The solid black line shows how applied current density changes over a batch as the diluate concentration is reduced. (e) The flexible operational domain of the current density for a voltage-controlled ED batch process with constant flow rate. The dashed lines show ratios of applied current density to limiting current density. (f) The flexible operational domain of the limiting current density for a voltage- and flow-controlled ED batch process. The solid black line show limiting current densities at varying flow rates. The shaded regions in each plot show the operational domains where a batch ED process could be operated. In b and c, an arbitrary trajectory of a variable power source is shown, with the corresponding applied current density trajectory to produce water shown in e and f, respectively. i_{lim} and i are limiting current density and applied current density, respectively. Q is flow rate. Q_{max} is the flow rate corresponding to maximum power utilization in voltage- and flow-controlled ED operation.

²¹² than the maximum possible.

Voltage-controlled ED creates an additional degree 213 of freedom in control by changing voltage applied to 214 the ED stack to manipulate the applied current density. 215 This functionality can be used to either maximize water 216 production by setting the current density always close 217 to limiting (as proposed by Shah et al. [25]), or to 218 maximize utilization of variable power by actively con-219 trolling the current density between zero and limiting. 220 In a voltage-controlled batch ED operation with a con-221 stant flow rate, the maximum ED power (associated with 222 the electrical field for removing, details in Section 3) is 223 224 determined by the ED operation at the highest current density, i.e. limiting current density; the pumping power 225 in a voltage-controlled ED system, as presented by Shah 226 et al., is constant. The flexible power range of this sys-227

tem is illustrated in Fig. 2b. Any power trajectory in 228 the flexible power range (Fig. 2b) can be met by vary-229 ing the current density during desalination via voltage 230 control (Fig. 2e). Although a voltage-controlled ED 231 system can be operated in a flexible domain, the applied 232 current density may be substantially lower than limiting 233 due to power restrictions imposed by a variable power 234 source (Figs. 2b and e). Therefore, voltage-controlled 235 operation may also underutilize the ED membranes to 236 produce water. 237

Under this control scheme, the flow controller would 244 optimally set the flow rate to set the appropriate limit-245 ing current density to fully utilize available power (as 246 illustrated by the dotted lines in Fig. 2f, details can be 247 found in Section 3). At the same time, the voltage con-248 troller would ensure the ED system was operating near 249 the "flow-controlled" limiting current density for fully 250 utilizing the membrane capacity. Therefore, voltage-251 and flow-controlled ED can always maximize water pro-252 duction rate while fully utilizing a variable power re-253 source. Furthermore, as illustrated in Fig. 2c), the 254 upper boundary of power consumption in voltage- and 255 flow-controlled ED can be much higher than in static 256 ED or voltage-controlled ED for a given system size, as 257 increasing flow rate increases limiting current density 258 and the power threshold. Note in Fig. 2c) that the power 259 domain can be reduced to zero by slowing the pumping 260 flow rate to zero. 261

262 263 Controlled ED batch operation

This section presents a control strategy in which wa-264 ter production and variable power utilization are co-265 maximized using two degrees of freedom in the ED 266 system - the voltage and flow rate. Water production 267 rate, which is dependent on desalination rate, is maximized by adjusting the voltage at each time step such 269 that the applied current density is maximized without 270 exceeding the limiting current density. Variable power 271 utilization is maximized by adjusting the flow rate at 272 each time step such that the power consumed closely 273 follows the power available from the source. 274

In our prior work, a robust ED static-operational model was proposed and validated [27]. This model parametrically describes the mass flow and power transfer between components (e.g., ED stack, pumps, etc.) and was demonstrated on multiple sizes of ED systems. This model is used herein to develop the time-variant ED control theory.

The static ED model is first discretized temporally 282 into multiple controlling time steps, τ_i , each of which 283 can be assigned a varying voltage and flow rate. At each 284 time step, the ED operation starts with a bulk diluate 285 concentration, $C_{d,0}^{b,\tau_i}$, and a bulk concentrate concentra-286 tion, $C_{c,0}^{b,\tau_i}$, at the point between each respective tank 287 and the ED stack inlets (illustrated in Fig. 3a). When 288 a voltage is applied, a concentration boundary layer of 289 thickness δ within a flow channel in the ED stack extends 290 from the membrane surfaces, where the concentration is 291 $C_{d/c,y}^{AEM/CEM,\tau_i}$, to the bulk flow, where the concentration 292

is $C_{d/c,y}^{b,\tau_i}$. The scripts *b*, *AEM*, *CEM*, *d*, and *c* designate bulk flow, the boundary layers near the AEM or CEM membrane, and the diluate or concentrate streams, respectively. The subscript *y* denotes the location along the discretized flow path, with *Y* the total discretized flow segments. 298

The ion increase/removal rate of concentrate/diluate ²⁹⁹ is controlled by varying the voltage, V^{τ_i} , and the flow ³⁰⁰ rates of the concentrate and diluate streams, $Q_c^{\tau_i}$ and $Q_d^{\tau_i}$, ³⁰¹ respectively: ³⁰²

$$\frac{(dC_{d,y}^{b})^{\tau_{i}}}{dt}^{\tau_{i}} = \frac{1}{NV_{y}^{cell}} \left[\mathcal{Q}_{d}^{\tau_{i}} (C_{d,y-1}^{b} - C_{d,y}^{b})^{\tau_{i}} - \frac{N\phi I_{y}^{\tau_{i}}}{zF} + \frac{NA_{y}D^{AEM} (C_{c,y}^{AEM} - C_{d,y}^{AEM})^{\tau_{i}}}{l^{AEM}} + \frac{NA_{y}D^{CEM} (C_{c,y}^{CEM} - C_{d,y}^{CEM})^{\tau_{i}}}{l^{CEM}} \right], \text{ and}$$
(1)

$$\left(\frac{dC_{c,y}^{b}}{dt}\right)^{\tau_{i}} = \frac{1}{NV_{y}^{cell}} \left[\mathcal{Q}_{c}^{\tau_{i}} (C_{c,y-1}^{b} - C_{c,y}^{b})^{\tau_{i}} + \frac{N\phi I_{y}^{\tau_{i}}}{zF} - \frac{NA_{y}D^{AEM} (C_{c,y}^{AEM} - C_{d,y}^{AEM})^{\tau_{i}}}{l^{AEM}} - \frac{NA_{y}D^{CEM} (C_{c,y}^{CEM} - C_{d,y}^{AEM})^{\tau_{i}}}{l^{CEM}} \right],$$
(2)

where V_y^{cell} is the volume of each segment, *z* is the ion charge, *F* is Faraday's constant (96,485 *C/mol*), *N* is the number of cell pairs, *I* is the current, ϕ is the current leakage factor, *A* is the membrane area, $D^{AEM/CEM}$ is the diffusion coefficient in the AEM and CEM membranes, respectively, and *l* is the thickness of membranes.

The diluate and the concentrate streams flow out from the ED stack and mix with the water in the diluate and concentrate tanks, respectively. The rate of concentration change in the diluate and concentrate tanks can be described as

$$\frac{dC_{d,0}^{b}}{dt})^{\tau_{i}} = \frac{Q_{d}^{\tau_{i}}}{V_{d}^{tank}} (C_{d,Y}^{b,\tau_{i}} - C_{d,0}^{b,\tau_{i}}), \text{ and } (3)$$

$$\left(\frac{dC_{c,0}^{b}}{dt}\right)^{\tau_{i}} = \frac{Q_{c}^{\tau_{i}}}{V_{c}^{tank}} (C_{c,Y}^{b,\tau_{i}} - C_{c,0}^{b,\tau_{i}}),$$
(4)

where $C_{d,0}^{b}$ and $C_{c,0}^{b}$ are the concentrations of the diluate and concentrate tanks (and ED stack inlets), respectively, and V_{d}^{tank} and V_{c}^{tank} are the volumes of the diluate and 316

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Figure 3: The illustrated flow of the diluate and the concentrate streams in an ED batch system with the associated electrical circuit model used to simulate time-variant ED operation. (a) The two streams in a batch ED system. For every control time step (τ_i) , $Q_d^{\tau_i}$, $Q_c^{\tau_i}$, $ad V^{\tau_i}$ represent the flow rate for the diluate, concentrate, and the voltage, respectively. The simulation model is described starting from the inlet of the stack to the outlet of the tanks, where the time step is updated (i = i + 1). C_d and C_c denote the concentration of the diluate and the concentrate, respectively. y = 1, 2, ..., Y, Y + 1 denote segment locations along the flow path. (b) The equivalent electrical circuit for the ED stack. The dashed line represents one segment (one value of y) and its equivalent circuit model. $R^{AEM,y}$ and $R^{CEM,y}$ are the area resistances associated with the AEM and CEM membranes, respectively. $V_{AEM,y}$ and $V_{CEM,y}$ are the potentials across the AEM membrane and the CEM membranes, respectively. $R_{d,y}$ and $R_{c,y}$ are the area resistances associated with the AEM and CEM membranes, respectively. I is the current flowing through the ED stack. I_{lim} is the current when the applied current density equals to the limiting current density.

concentrate tanks, respectively. The desalination rate of
the ED system is the desalination rate of the diluate tank,
given by Eq. 3.

To calculate the total current, the ED stack is modeled as an analogous DC circuit (Fig. 3b), with a current flowing through each discretized segment

$$I_{y}^{\tau_{i}} = \phi_{A}(\frac{WL}{Y})i_{y}^{\tau_{i}},$$
(5)

where *W* is the stack width, *L* is the membrane channel length, ϕ_A is the open area porosity of the turbulencepromoting channel spacer, and *i* is the current density. The equivalent circuit elements for each discretized segment are connected in parallel, and thus the voltage is equal across all segments.

To maximize the desalination rate at a given flow rate, the segment current should be maximized, as indicated by Eq. 1. The limiting current density determines the maximum applied current density that can be supported by the ED system before splitting water occurs, as discussed in Section 2. It can be approximated as a function of the bulk diluate concentration with 335

$$r_{lim,y}^{+,-} = \frac{zFkC_{d,y}^{b}}{t^{AEM,CEM} - t^{+,-}},$$
 (6)

where $t^{+,-}$ is the minimum of the dimensionless anion (-) and cation (+) transport numbers in the bulk solution, $t^{AEM,CEM}$ are the transport numbers of the AEM and CEM membranes, respectively, and k is the mass transfer coefficient. k can be represented as

$$k = \frac{ShD_{aq}}{d_h},\tag{7}$$

where D_{aq} is the diffusion coefficient of the aqueous solution, d_h is the hydraulic diameter, and Sh is the Sherwood Number. Sh represents the mass transfer performance, and is correlated with the Reynolds number and the Schmidt number. These relationships are described further in Appendix A. 342 As indicated by Eq. 6, the limiting current density is proportional to the bulk diluate concentration. As the voltage increases, the applied current density of the last segment (y = Y = 5) is the first to reach the limiting current density because the bulk diluate concentration at the outlet is the lowest within the ED stack. Thus, the maximum voltage that can be applied without exceeding the limiting current density is the voltage when the applied current density of the last segment is close to the limiting current density. This maximum voltage is

$$V^{\tau_{i}} = V_{el} + N(V_{Y}^{CEM} + V_{Y}^{AEM}) + Nr_{i}i_{lim,Y}(R_{d,Y} + R_{c,Y} + R_{Y}^{BL} + R^{AEM,Y} + R^{CEM,Y}),$$
(8)

where: V_{el} is the electrode potential (1.4 V when hydro-347 gen ions are reduced at the cathode and chloride ions are 348 oxidized at the anode); VCEM,Y, VAEM,Y are the poten-349 tials across the CEM and AEM membranes, respectively; 350 R_{Y}^{BL} , $R^{AEM,Y}$, $R^{CEM,Y}$ are the area resistances associ-351 ated with the concentration boundary layers, the AEM 352 membranes, and CEM membranes, respectively; and r_i 353 is the safety factor for approaching the limiting current 354 density. r_i provides an additional degree of freedom to 355 track (with an appropriate safety-margin) the limiting 356 current density throughout the batch process [26]. $R_{d,Y}$ 357 and $R_{c,Y}$ are the resistances associated with the dilu-358 ate and concentrate streams, respectively, which can be 359 further represented as 360

$$R_{d,Y} = R_{d,Y}^b + R_{d,Y}^{AEM} + R_{d,Y}^{CEM}$$
, and (9)

$$R_{c,Y} = R_{c,Y}^b + R_{c,Y}^{AEM} + R_{c,Y}^{CEM},$$
 (10)

where $R_{d/c,Y}^{b}$ is the resistance of the bulk flow, and $R_{d/c,Y}^{AEM}$ and $R_{d/c,Y}^{CEM}$ are the resistances in the boundary layers near the membrane surfaces, respectively. These equivalent resistances depend on the diluate and concentrate concentrations; detailed derivations for these resistances can be found in Wright et al. [27].

The potentials associated with the concentration difference across the exchange membranes, $V_{AEM,Y}$ and $V_{CEM,Y}$, can be approximated by

$$V_{AEM,Y} = \frac{(2t^{AEM} - 1)RT}{F} \log(\frac{\gamma_c C_{c,Y}^{AEM}}{\gamma_d C_{d,Y}^{AEM}}), \text{ and } (11)$$

$$V_{CEM,Y} = \frac{(2t^{CEM} - 1)RT}{F} \log(\frac{\gamma_c C_{c,Y}^{CEM}}{\gamma_d C_{d,Y}^{CEM}}), \quad (12)$$

where *T* is the temperature and R is the gas constant, $8.31JK^{-1}mol^{-1}$ [27].

To maximize variable power utilization, total system 372 power consumption is adjusted to closely follow the input 373 power. The total system power consumption of a timevariant ED system is estimated by summing the power 375 consumption of the most power-consuming components, 376 which are the DC power supply for the ED stack and the 377 diluate and concentrate pumps: 378

$$P_{total}^{\tau_i} = P_{ED}^{\tau_i} + P_{pump,d}^{\tau_i} + P_{pump,c}^{\tau_i}, \qquad (13)$$

where $P_{pump,d}$ and $P_{pump,c}$ denote the power consumed by the diluate pump, and the concentrate pump, respectively. P_{ED} denotes product of the voltage and current applied to the ED stack. 382

The power consumed by the variable speed pumps of the diluate and concentrate streams will depend on the flow rate and the hydraulic characteristics of the full ED system. In general, the power consumption of a variable speed-controlled centrifugal pump follows the Affinity Laws (also known as "the Pump Laws") [28], 383

$$\frac{Q_{d/c}^{\tau_i}}{Q_{ref}} = \frac{n_{d/c}^{\tau_i}}{n_{ref}},\tag{14}$$

$$\frac{H_{d/c}^{\tau_i}}{H_{ref}} = \left(\frac{n_{d/c}^{\tau_i}}{n_{ref}}\right)^2, \text{ and}$$
(15)

$$\frac{P_{pump,d/c}^{\tau_i}}{P_{ref}} = (\frac{n_{d/c}^{\tau_i}}{n_{ref}})^3,$$
(16)

where *n* is pump speed and *H* is the pump head. Q_{ref} , H_{ref} , P_{ref} , and n_{ref} indicate the referenced operation points of the system.

The power consumption of the DC power supply in an ED stack (i.e. the desalinating power) is estimated as the product of the current and the applied voltage, 394

$$P_{ED}^{\tau_i} = (VI)^{\tau_i}.$$
 (17)

To match the instantaneous power input, the instantaneous power consumption of the ED system, $P_{total}^{\tau_i}$, is controlled by varying the voltage and flow rate. As shown by Eq. 14 and Eq. 16, the pumping power explicitly depends on the flow rate, which can be used to estimate the new pumping power when a new flow rate is applied. 401

To estimate the new desalinating power, $P_{ED}^{\tau_i}$, when a new voltage is applied to the ED stack is non-trivial. It requires summing all of the segments' currents, as shown in Fig. 3, which requires solving a system of equations, 402

including Eq. 1 and Eq. 2, at varying flow rates. How-406 ever, variable power inputs from solar or wind sources, 407 or changes in electricity tariffs in dynamic grid pricing, 408 may vary on the order of seconds, requiring the con-409 troller to respond quickly to identify and apply optimal 410 voltages and flow rates. To accelerate the controller's 411 computational efficiency, an explicit method of estimat-412 ing the ED desalination power under varying flow rate 413 conditions is proposed. 414

To reduce computation time, the controller only considers electromigration for ion transfer. Electromigration generally contributes $\geq 90\%$ of the mass transfer in ED desalination [27, 29], and the contribution is even higher with a high current (enabled by a high flow rate, as indicated by Eq. 6). This assumption results in the explicit current estimation

$$I_{appr}^{\tau_i} = \frac{Q_d^{\tau_i} (C_{d,0}^b - C_{d,Y}^b)^{\tau_i} zF}{N\phi},$$
 (18)

where $I_{appr}^{\tau_i}$ is the approximated current. In this case, the transience of the changing flow rate is negligible compared to the transience of the changing dilute concentration, due to the incompressible nature of water.

Using the approximated current (Eq. 18) and the maximized voltage (Eq. 8), the ED desalination power at the new flow rate can be explicitly estimated (Eq. 17). The total power consumption can then be evaluated at a different flow rate and combined with Eq. 16, which enables the controller to efficiently optimize flow rates to match or closely follow the available variable power input.

Figure 4 shows a flowchart of the final model-based 434 controller, incorporating both desalination rate and 435 power utilization components. By using this controller, 436 water production is maximized by setting a voltage that 437 maximizes the ion transfer rates and avoids water split-438 ting occurring in the stack at a particular flow rate. Then 439 the variable power utilization is maximized by setting the 440 flow rate using an optimization feedback loop that min-441 imizes the difference between the power consumption 442 and the available power input. As a result, this strategy 443 enables the simultaneous maximization of water produc-444 tion and variable power utilization, facilitating the most 445 efficient use of water and available power at every point 446 in time. 447



Figure 4: Flowchart of the model-based controller for time-variant ED operation. $P_{total}^{\tau_i}$ and $P_{input}^{\tau_i}$ are the total power consumption (including the ED power and the pumping power) and the variable power input at every control time instant τ_i , respectively. Y refers to the total discretized sections of the ED stack referenced in Fig. 3.

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4. Pilot time-variant ED system design

4.1. Experimental setup

A pilot-scale time-variant ED prototype was built to 450 validate the proposed control theory, following the con-451 figuration shown in Fig. 5. The ED stack was manufac-452 tured by Suez Water Technologies and Solutions (Model 453 AO3-1-2-50-35), with the parameters listed in Table 1. 454 Two pumps (Xylem Goulds 3SV-11) recirculated the 455 diluate and concentration streams with their speed con-456 trolled by pump controllers (Xylem CentriPro Aquavar). 457 A 60-25V DC supply (TDK-Lambda GEN) supplied 458 the voltage (regulated to $\pm 1\%$ of the commanded value). 459 The polarity of the applied voltage was reversed between 460 batches by switching the diluate and concentrate chan-461 nels in the stack using valves. This reversal operation 462 has been shown to reduce the scaling propensity in ED 463 desalination [30]. 464

Two flow meters (Omega FP1408) were used to mon-465 itor the flow rate $(\pm 1\%)$ in the diluate and concen-466 trate streams. In-line conductivity probes (Connectiv-467 ity Instruments CDCE-90) interfacing with conductivity 468 controllers (Connectivity Instruments CDCN-91) moni-469 tored the conductivity (to an accuracy of $\pm 2\%$) at the en-470 try and exit of the ED stack. All sensors interfaced with 471 a CLICK I/O Programmable Logic Controller (PLC) 472 with analog input and output modules (C0-04AD-1, C0-473 04AD-2, and C0-04DA-2). Each electrode was rinsed 474



Figure 5: Major system elements and their interactions for the time-variant ED prototype tested at the Brackish Groundwater National Desalination Research Facility (BGNDRF) in Alamogordo, New Mexico.

Design Variables	Value
ED cell pairs	30
Diluate tank volume, m ³	0.42 ±4%
Brine tank volume, m ³	0.28 ±7%
Flow Path Width, cm	19.7
Flow Path Length, cm	168
AEM Resistance, Ωcm^2	7
CEM Resistance, Ωcm^2	10
Void fraction	0.83±0.03
Area porosity	0.70±0.02
Spacer thickness, mm	0.71±0.01

Table 1: Parameters of the ED stack

with a sodium sulfate solution (conductivity over 14 mS/cm $\pm 2\%$) held at a flow rate of 6-8 LPM ($\pm 1\%$).

Feed water was taken from Well No. 1 at the Brackish Groundwater National Desalination Research Facil-

479 ity (BGNDRF) in Alamogordo, New Mexico. Major

constituents in the water are listed in Table 2. The feed 480 water salinity was similar to that of a previous pilot-scale 481 PV-ED field study conducted by our group in rural In-482 dia [12]. Water quality measurements were performed 483 by DHL Laboratories (San Antonio, TX). In each batch 484 reported in the following section, the feed water was de-485 salinated to a target product concentration of 500 μ S/cm 486 with a batch size of 0.42 m^3 . 487

4.2. Controller implementation

The pump speeds and voltage applied to the ED stack 489 electrodes were controlled by variable frequency drives 490 (VFDs) and a programmable DC power supply accord-491 ing to the received control signals from the implemented 492 controller script, respectively. The controller strategy, 493 implemented in Python, calculated an optimal voltage 494 and flow rate using real-time measurements of variable 495 power inputs and conductivity from both the diluate and 496 concentrate streams, based on the model introduced in 497

Parameters	Value	
Na^+ , mg L ⁻¹	293±29	
$Mg^{2+}, \mathrm{mg} \mathrm{L}^{-1}$	12.6±1.3	
$Ca^{2+}, \mathrm{mg} \mathrm{L}^{-1}$	54.6±5.5	
Cl^{-} , mg L ⁻¹	38.1±3.8	
$SO_4^{2-}, \text{mg} \text{L}^{-1}$	504±50	
Alkalinity Bicarbonate, $mg L^{-1}$ as $CaCO_3$	161±1	
Total dissolved solids (TDS), $mg L^{-1}$	995±72	
Conductivity, $\mu S cm^{-1}$	$1,500\pm30$	

Table 2: The major constituents in the brackish groundwater from Well NO.1 at the Brackish Groundwater National Desalination Research Facility (BGNDRF), measured on 3-Dec-2018.

Section 3. Pump performance curves were experimentally generated from the two installed pumps based on measurements at multiple speeds. Speed versus flow and speed versus power pump curves were empirically fit to the experimental data and used in the controller implementation, as described in Section 3. The fitted pump curves are plotted in Appendix B.

Control signals were applied to the time-variant ED 505 prototype in an open loop. Communication between 506 the controller script and modules in the prototype was 507 implemented via PLC modules. Using measured con-508 centrations at the current time, controller predictions 509 were used to optimize the flow rate and voltage. Sig-510 nals for these values were then sent to the VFDs and the 511 DC power supply to control the flow rate and voltage, 512 respectively, for the upcoming time step. The duration 513 of the time step was 3 s, based on preliminary test-514 ing and chosen to capture variations in the power input 515 while allowing enough time for the ED system to reach 516 a new steady state after the latest change in voltage and 517 flow rate. The system response time was determined 518 experimentally. The same time step of 3 s was used for 519 simulation studies. 520

5. Pilot time-variant ED system testing and results

522 5.1. Controller test for variable voltage, high constant 523 flow rate ED

The efficacy of the control theory presented in Section 3 was first tested with a fixed, high flow rate and variable voltage to see if the controller could produce an increased desalination rate compared to static ED operation. The maximum flow rate in an ED system depends on many factors and is determined by the maximum pump speed. In this study, the maximum linear velocity in the membrane channels was restricted to be ~ 20 cm/s 531 $(\pm 1\%)$, corresponding to 42 LPM bulk flow rate. This 532 is already significantly higher than the velocity in the 533 membrane channels of conventional static ED operation 534 (4-12 cm/s) [25], which is set to ensure operational sta-535 bility of the membranes and spacers. Figure 6 shows 536 the current, power consumption, and diluate conductiv-537 ity over a batch for voltage-controlled ED operation at 538 the maximum flow rate of 42 LPM. Static ED operation 539 with a flow rate of 25 LPM (~12 cm/s in the membrane 540 channels) is shown for comparison. In these tests, the 541 current density was not allowed to exceed 70% of limit-542 ing (which is a function of flow rate, described in Section 543 3).

Figure 6 demonstrates that running the time-variant 545 ED prototype at a high flow rate increased the opera-546 tional domain (shaded region) compared to the single 547 operating trajectory for static ED (black line), achieving 548 a 45% increase in desalination rate (Fig. 6c). Further 549 comparisons of static ED to time-variant ED at different 550 flow rates are shown in Appendix C. The controller pre-551 dictions and the experimentally measured current and 552 power were consistently aligned (within 1.5% RMS er-553 ror). This indicates that the controller can accurately 554 predict current and then successfully predict power con-555 sumption. The results in Fig. 6 show that the controller 556 could enable the time-variant ED system to directly use 557 variable power sources over a wide range of operating 558 conditions by varying flow rate and ED stack voltage, as 559 discussed in Section 3. 560

5.2. Controller test for voltage- and flow-controlled ED 561

To further characterize the performance of voltage-562 and flow-controlled ED operation, and validate the con-563 trol theory presented in Section 3, the time-variant ED 564 prototype system was run using a representative solar 565 power input profile (Fig. 7a) during one batch. The 566 solar profile was recorded by a set of local solar panels 567 (Hyundai HiS-S285RG) at BGNDRF. The target prod-568 uct concentration was set to 300 mgL⁻¹ (~500 μ S cm⁻¹) 569 for this test. The controller was able to command the 570 prototype ED system to consumer power on a trajectory 571 that closely followed (within 10.0% RMS error) the ref-572 erence solar power profile (Fig. 7a) while maintaining 573 a measured product water concentration of 300 mgL⁻¹. 574 These results demonstrate the ability of the time-variant 575 ED system to adaptively desalinate water to a desired 576 product concentration while adjusting voltage and flow 577 rate to match an arbitrary variable power level. This flex-578 ibility could allow the time-variant ED system to directly 579 integrate with real clean energy sources, such as solar or 580 wind, without requiring significant battery capacity. 581



Figure 6: Controller predictions and experimental ED batch performance under voltage-controlled variable voltage (VV) and high constant flow rate (CQ) (42 LPM) conditions versus constant voltage (CV), moderate constant flow rate (CQ) (25 LPM) conditions. Results are presented for: (a) current versus diluate conductivity; (b) total power of the ED system versus diluate conductivity; and (c) diluate conductivity versus batch time (experimental performance only). The shaded regions in (a) and (b) represent the flexible operational domain for which a flexible ED system could operate using direct power from a variable power source.

Figure 7b shows the controlled power consumption 582 profile of the pilot-scale time-variant ED system while 583 the controller was fed three arbitrary constant input 584 power levels of 1000 W, 850 W, and 730 W. Constant 585 power levels were chosen as inputs for three primary 586 reasons. First, any variable power source can be approx-587 imated as constant for a very short duration. Second, 588 at each controlling time step (every 3 s in these ex-589 periments), the controller needs to optimize and adjust 590 voltage and flow rate to match a singular power value; 591 whether this value changes in time or not is arbitrary in 592 the perspective of the controller. To maintain a constant 593 power consumption, the controller has to continuously 594 make adjustments, just as it would to follow a variable 595 input power profile. Third, operating a constant power 596 while maximizing water production rate simulates real-597 world situations where power consumption would have 598 to be maintained under a threshold, say within the speed 599 limitations of a wind turbine or in an industrial grid-600 powered application where there are different charge 601 rates depending on power draw. Therefore, the three 602 constant power levels were used to robustly test the load 603 flexibility of the prototype time-variant ED system and 604 demonstrate the utility of the control model. 605

The results in Fig. 7b demonstrate that the controlled 606 time-variant ED system power consumption was able to 607 608 closely match the predefined constant input power levels for all three cases; the RMS errors for each test were 609 1.7% for 1000 W, 4.2% for 850 W, and 6.3% for 730 W, 610 as the batch desalinates from 1400 μ S/cm to about 500 611

 μ S/cm. All of the time-variant ED operations had higher 612 desalination rates (ranging from 6-15%) than the static 613 ED process used as a benchmark (Fig. 7c). The shaded 614 regions in Figs. 7a and b show how much flexibility 615 remains in the operational domain, with the upper limit 616 bounded by the same maximum power conditions shown 617 in Fig. 6, defined by variable voltage operation and a 618 constant pumping flow rate of 42 LPM. 619

5.3. Desalinating and pumping power

Although the time-variant ED batch trajectories 621 largely align with their respective input power profile, 622 there are some small deviations. Particularly in the 623 cases with relatively low power input, the measured ex-624 perimental power tends to fluctuate around the variable 625 power input. To explore this deeper, the data from Fig. 626 7b were decomposed to analyze the power contributions 627 from the ED desalination process and pumping.

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Figure 8 shows that the measured and predicted ED 629 desalination power values follow the same trends for the 630 three tested cases. However, for power levels of 850 631 W and 730 W, the measured total desalination power 632 is slightly under the predicted power. These deviations 633 are likely due to the neglected back diffusion through 634 the membranes into the channels; because the controller 635 does not take back diffusion into account, it tends to 636 predict a higher current for a given applied voltage (Eq. 637 18). The power becomes increasingly over-predicted at 638 lower flow rates because the back diffusion is higher, as 639 seen in the 850W and 730W cases. 640



Figure 7: Time-variant ED system performance for varying power inputs. (a) Measured power usage for time-variant voltage- and flow-controlled ED operation from the pilot-scale ED prototype while following a representative, measured solar power profile during one batch. (b) Measured power consumption for time-variant voltage- and flow-controlled ED operation from the pilot-scale ED prototype under three constant power inputs (1000 W, 850 W, and 730 W) during one batch. (c) The corresponding conductivity profiles from the results in (b). A benchmark constant voltage, constant flow rate static ED batch (CVCQ) process at 25 LPM flow rate is shown for comparison in (b) and (c). To demonstrate the operational limits at maximum pumping power, a variable voltage, constant flow rate ED batch process (VVCQ) at 42 LPM is shown in (a) and (b), which marks the upper boundary of the operational domain (shaded region).

The measured pumping power follows the general 641 trend of the predicted pumping power in all of the tested 642 cases, with the exception of some small fluctuations in 643 the 850 W and 730 W cases (Figs. 8b and c, respec-644 tively). The small fluctuations at lower input power 645 levels may be caused by the behavior of the pump when 646 operating outside its intended performance curve. The 647 applied voltage and flow rate in the three cases are plot-648 ted in Figs. 9a and b, respectively. The pumps used in 649 the prototype ED pilot have their highest efficiency at 650 flow rates of 40-70 LPM. Figure 9b plots the flow rate 651 for each input power level during the ED batch. The 652 pumps were operated in a region outside their intended 653 performance curve for the lower power levels of 850 W 654 and 730 W, where they would be expected to perform 655 less predictably and stably. They were operated closer 656 to their high efficiency operation zone at the power level 657 of 1,000 W. A slightly downsized pump may have im-658

proved the power fluctuations seen in Fig. 8. In spite of these small fluctuations, the measured pumping power closely followed the values predicted by the controller (to within 1.8% RMS for 1000 W, 4.6% RMS for 850 W, and 5.7% RMS for 730 W, as the batch desalinates from 1400 μ S/cm to about 500 μ S/cm).

A large spike in the voltage is apparent in Fig. 9a at 665 the beginning of each batch for all three power levels, 666 accompanied by a rapid drop in flow rate just before the 667 voltage spike. These features are caused by an under-668 prediction of current at the beginning of the batch by 669 the controller. At this moment, the electric potential is 670 instantaneously applied across the membranes in the ED 671 stack. In a real stack, for the initial diluate concentra-672 tion in the membrane channels to be perturbed by the 673 electrical field, salts must accumulate before building 674 up concentration variations between the inlet and outlet. 675 These transient effects are ignored by the controller, and 676



Figure 8: Experimentally measured power consumption of the pump and the ED desalination process, and the controller predictions for reference power inputs of (a) 1000 W, (b) 850 W, and (c) 730 W.

therefore, the approximated current from Eq. 18 underestimates the applied current at this moment, causing
an overprediction of flow rate in the first few instants,
as seen in Fig. 9b. After the voltage spike, the concentration drop across the diluate stream becomes fully
developed and the effect of the accumulating salts be-

comes insignificant.During this transient period, the683control model has a large error (compared to the period684after the spike) due to the assumptions used in the model685being briefly invalid.Therefore, for this short duration,a small amount of battery energy storage is required to687supply enough power.This transient period is generally



Figure 9: Experimentally measured voltage and flow rates during time-variant ED operation with reference power inputs of (a) 1000 W, (b) 850 W, and (c) 730 W.

very short (less than 1 min per 25-40 min batch in the
 pilot system), and the required battery capacity is nearly
 negligible. For example, for the pilot time-variant sys tem presented herein that requires batteries for reshaping
 power over ~1 min, the required battery capacity could
 be as small as 1/40-1/25 the required battery capacity
 of traditional renewable-powered ED systems that use

batteries to reshape the variable power input throughout 696 the batch. 697

5.4. The trade-off between production rate and energy consumption 698

Although voltage and flow rate follow the same trends 700 in variation across the three input power levels shown in 701 Fig. 9, they differ in magnitude. Higher power lev-702 els tend to have higher flow rates and higher voltages. 703 Comparing pumping power consumption in Fig. 8 with 704 ED stack voltage in Fig. 9a, the pumping power varies 705 significantly with varying voltage in each of the three 706 power levels, but the measured ED desalination power 707 does not. This indicates that the scaling factors for flow-708 to-power and voltage/current-to-power differ. Because 709 the feed concentration (~1500 μ S cm⁻¹) in each case 710 is desalinated to a same product concentration (~500 711 μ S cm⁻¹), the electrical resistances (Eqs. 9 and 10) of 712 both the concentrate and the diluate streams should be 713 similar, independent of case. As a result, the ED de-714 salination power primarily scales with V^2 , according to 715 Eq. 17. In contrast, the pumping power scales with Q^3 , 716 according to Eq. 16. As a result, the pumping power in-717 creases much faster with flow rate, causing the pumping 718 to consume more power than ED in all three test cases. 719 Efficient pumping is therefore critical to improve the 720 energy efficiency of time-variant ED batch operation. 721

Table 3 gives the specific energy consumption (SEC) 722 and desalination rate for the three time-variant, voltage-723 and flow-controlled ED batch cases at different power in-724 puts, along with a static ED batch process with a flow rate 725 25 LPM. The starting feed concentration was slightly dif-726 ferent from batch-to-batch during the experiments; each 727 was run with a target product concentration of 500 ± 5 728 μ S cm⁻¹. The results in Table 3 indicate the trade-off 729 between SEC and batch time (equivalent to desalina-730 tion rate in m^3/h) and suggest they are correlated non-731 linearly. The desalination rate was increased by 29%, 732 20% and 19% by using 62%, 52% and 30% more en-733 ergy, respectively, compared to static ED. The pumping 734 SEC also corroborates the significantly increased frac-735 tion of energy consumed by pumping compared to ED 736 desalination under higher input powers. The percentage 737 contribution of pumping to SEC increases from 68% in 738 the static ED case to 74% in the time-variant ED case 739 with input power of 1,000 W. 740

Parameters	CVCQ@25LPM	VVVQ@730W	VVVQ@850W	VVVQ@1000W
Feed concentration $[\mu S/cm]$	1340±27	1460 ± 29	1560±31	1410±28
Product concentration $[\mu S/cm]$	500±10	505±10	500±10	500±10
Batch time [min]	35.9±3.3‰	29.0±3.3‰	28.9±3.3‰	25.5±3.3‰
SEC $[kWh/m^3]$	0.63±0.03	0.82 ± 0.04	0.96 ± 0.05	1.02 ± 0.05
Pumping SEC $[kWh/m^3]$	0.43 ± 0.02	0.56 ± 0.03	0.68 ± 0.04	0.75 ± 0.04
ED desalination SEC [kWh/m^3]	0.20 ± 0.01	0.26 ± 0.01	0.28 ± 0.01	0.27±0.01
Batch time compared to CVCQ	N/A	82%±5.8‰	81%±5.8‰	71%±5.8‰
SEC compared to CVCQ	N/A	130%±6.8%	150%±7.0%	160%±6.8%
Pumping SEC compared to CVCQ	N/A	130%±7.1%	160%±7.5%	170%±7.1%
ED desalination SEC compared to CVCQ	N/A	130.00%±6.3%	140.00%±6.1%	130.00%±6.2%

Table 3: Performance of the pilot-scale prototype in variable voltage, variable flow (VVVQ), time-variant ED batch operation with constant power inputs of 730W, 850W, and 1000W. Performance of a benchmark constant voltage, constant flow (CVCQ), static ED batch process at a flow rate of 25LPM is given for comparison. All tests were run with a target product concentration of $500\pm5 \,\mu\text{S cm}^{-1}$.

741 6. Discussion

In this work, the time-variant method for operating 742 a batch ED system has been demonstrated to be load-743 flexible and able to accommodate variable power sources 744 and maximize the rate of water production, in order to 745 reduce the water cost. This new operational strategy is 746 analogous to how multi-stage ED stacks, or series as-747 semblies of ED stacks, are arranged with different volt-748 ages applied to each electrical stage to maintain applied 749 current density near limiting, and different numbers of 750 parallel flow channels to manipulate flow velocity for 751 a desired limiting current density and/or to minimize 752 pumping power [31]. Because time-variant ED batch 753 operation is able maintain applied current density near 754 limiting and fully utilize an available power source for 755 maximized water production rate, ED systems designed 756 with this technology may result in reduced capital costs 757 compared to static, continuous ED systems composed of 758 multi-stage ED stacks or multiple ED stacks in series. 759 The hardware required to make a time-variant ED system 760 is readily available off-the-shelf, with some components 761 (e.g. conductivity sensors) already routinely included in 762 conventional ED batch systems. 763

Section 5.3 reveals a trade-off between power con-764 sumption and desalination rate. Flexible operation al-765 lows ED systems to utilize much higher levels of power 766 compared to a similarly sized static ED system, which 767 will increase desalination rate but result in higher SEC. The additional energy consumption of the accelerated 769 production rate may not be an issue in some applica-770 tions in which operational time is critical, or available 771 772 energy is abundant (e.g. solar irradiance at mid-day). To be economically viable therefore, the cost of energy 773 provided by either on-grid or off-grid sources should be 774 low enough to justify the additional energy consumption 775

required to operate time-variant ED systems at the high flow rates required for maximizing desalination rate. For on-grid cases, operation costs could be reduced while maximizing production rate by either limiting the overall system power threshold and/or the pumping power threshold.

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To justify higher SEC in on-grid applications, time-782 variant ED technology could utilize variable electricity 783 tariffs between peak and off-peak times, which are part 784 of a demand response (DR) approach for reducing peak 785 loads [32]. Various DR programs provide financial ben-786 efits to customers who are willing to shift loads from 787 peak times to off-peak times. Wang and Li [16] surveyed 788 time-of-use pricing services in the US and found that 789 peak time prices can be 500-600% higher than off-peak 790 time prices in summer months (June-September), and 791 30%-200% higher in other months. Such considerable 792 price differences could incentivize the adoption of time-793 variant ED systems to produce more water, or produce 794 water at a faster rate during off-peak periods, than exist-795 ing technologies, which could potentially reduce overall 796 water costs. Time-variant ED systems could also facili-797 tate the integration of renewable energy sources into the 798 electrical grid by providing a consumer of excess energy 799 production on an irregular schedule, thereby reducing 800 carbon emissions from energy sources currently used to 801 meet peak demands (e.g. coal and natural gas). 802

For off-grid applications, time-variant operation could 803 enable ED systems to directly utilize all available inter-804 mittent renewable energy, such as peak midday solar 805 irradience that would otherwise be neglected or stored 806 in batteries. This could significantly reduce system cap-807 ital costs by reducing the battery capacity required for 808 renewable energy peak shifting. Small battery capac-809 ity and high water production rates would be particu-810 larly valuable for disaster response applications, where 811

small-scale, lightweight, PV-powered time-variant ED 812 systems could be rapidly shipped and deployed. For 813 microgrid solar systems, which are gaining popularity 814 in cost-constrained, remote communities in developing 815 countries [33], time-variant ED systems could reduce 816 electricity costs by utilizing otherwise unused solar en-817 ergy and creating additional value through the produc-818 tion of potable water. 819

7. Conclusions 820

This paper proposes a highly-flexible and production-821 optimized ED desalination technology for brackish wa-822 ter with two degrees of freedom of control: applied 823 ED stack voltage and pumping flow rate. This control 824 method can enable flexible and effective uses of variable 825 power sources on a timescale of seconds to maximize 826 water production, which has particular value in utilizing 827 renewables (e.g. wind and solar). Additionally, time-828 variant ED operation can improve utilization of mem-829 brane area by maximizing the applied current density, 830 which could facilitate smaller and lower-cost desalina-831 tion systems to hit a target production volume, compared 832 to what can be achieved with static ED operation. 833

A pilot-scale, time-variant ED system was designed 834 and built to validate the theory presented in this work. 835 The time-variant system was able to utilize up to ~ 3 more 836 power than if operated at static voltage and flow rate, 837 achieving up to 45% greater desalination rates. Within 838 the operational domain, the pilot system was shown to 839 successfully operate at three different power inputs, suc-840 cessfully adjusting voltage and flow rate as anticipated. 841 A trade-off between SEC and desalination rate was iden-842 tified; in the three tests with different power levels, de-843 salination rate was increased by 29%, 20% and 19% by 844 using 62%, 52% and 30% more energy, respectively, 845 compared to static ED batch operation. 846

For on-grid applications, time-variant ED operation 847 could enable water producers to align production time 848 and power consumption favorably with energy tariffs, 849 which are lower in the evening. For off-grid systems, 850 time-variant ED could remove or reduce the need for 851 batteries (and their associated costs) by producing water when energy is available. The technology presented 853 herein may enable engineers to design brackish water 854 ED desalination systems for new applications, smaller 855 856 size scales, and at lower costs than what can be achieved with current technology. As a result, time-variant ED 857 may have particular value as a potable water source for 858 poor, off-grid communities in developing countries. 859

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9. Appendix

9.1. Appendix A: hydraulic diameter and the Sherwood number 1009

In the mass transfer coefficient k, the hydraulic diameter d_h is

$$d_h = \frac{4\epsilon}{2/h + (1-\epsilon)(8/h)},\tag{19}$$

where ϵ is the void fraction. The Sherwood Number, a measure of mass transfer performance, is correlated to the Reynolds Number and the Schmidt number by

$$Sh = aRe^b Sc^c. (20)$$

The Schmidt number Sc is a material dependent, non-1010 dimensional quantity relating the momentum and mass 1011 diffusion. The Reynolds number is a dimensionless 1012 number relating inertial to viscous stresses in the flow. 1013 They are 1014

 $Sc = \frac{\mu}{\rho_{aq} D_{aq}}$

$$Re = \frac{\rho_{aq} u_{ch} d_h}{\mu}.$$
 (22)

9.2. Appendix B: The pump curve used in this paper

Two pumps were used in the ED system presented: a 1016 diluate pump and a concentrate pump. The two pumps 1017 were the same model, but the performance slightly varied 1018

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(21)

due to differences in their associated hydraulic circuits 1019 in the ED system. In order to mitigate the fouling, elec-1020 trodialysis reversal (EDR) operation was used during 1021 testing, such that the polarity of the electrical field was 1022 reversed after each batch. As a result, each pump oper-1023 ated in two positions, namely position 1 and position 2. 1024 Figure 10 presents the experimentally measured pump 1025 curves, which were used to predict pump performance 1026 in this work. 1027



Figure 10: Pumps curves of the two pumps used in the pilot time-variant ED system.

9.3. Appendix C: Desalination rate of CVCQ at different 1028 flow rates 1029

An appropriate flow velocity is determined by the 1030 trade-off between pumping power and ED stack power 1031 consumption, which is expected to be small enough to 1032 reduce pumping power, but just high enough to increase 1033 the limiting current density and to limit concentration 1034 polarization [34]. Consequently, the flow rate of a con-1035 ventional ED batch is usually between 4-10 cm/s in each 1036 membrane channel according to prior experimental and 1037 theoretical studies [35, 36, 37], and the manufacture's 1038 recommended flow rate of (~7 cm/s) [27]. Therefore, 1039 Fig. 11 presents the desalination performance of static 1040 ED operation with several flow rates for potential com-1041 parisons. 1042



Figure 11: The diluate conductivity versus the batch time of CVCQ operation at various flow rates.

Acronyms	1044
AEM Anion exchange membranes.	1045
BGNDRF Brackish Groundwater National Desalina- tion Research Facility.	1046 1047
CapEx Captial expenditure.	1048
CEM cation exchange membranes.	1049
CVCQ Constant voltage constant flow rate.	1050
DC Direct current.	1051
DR Demand response.	1052

- 1053 ED Electrodialysis.
- ¹⁰⁵⁴ **OpEx** Operational expenditure.
- ¹⁰⁵⁵ **PLC** Programmable logic controller.
- 1056 **PV** Photovoltaic.
- 1057 **RO** Reverse osmosis.
- ¹⁰⁵⁸ **SEC** Specific energy consumption.
- 1059 **TDS** Total dissolved solids.
- ¹⁰⁶⁰ **VFD** Variable frequency drive.
- ¹⁰⁶¹ **VVCQ** Variable voltage constant flow rate.
- ¹⁰⁶² **VVVQ** Variable voltage variable flow rate.

1063 Symbols

- 1064 *A* Membrane area, m²
- C_c^{b} Bulk concentration of concentrate, mol m⁻³
- ¹⁰⁶⁶ C_d^b Bulk concentration of diluate, mol m⁻³
- $_{1067}$ d_h Hydraulic diameter, m
- ¹⁰⁶⁸ D Diffusion coefficient, m² s⁻¹
- ¹⁰⁶⁹ F Faraday constant, 96485 C mol⁻¹
- 1070 H Pump head, m
- $_{1071}$ *i* Current density, A m⁻²
- 1072 I Current, A
- ¹⁰⁷³ k Mass transfer coefficient, m s⁻¹
- L Membrane channel length, m
- $_{1075}$ N Number of cell pairs
- 1076 P Power, W
- ¹⁰⁷⁷ Q_c Flow rate of concentrate, m³ s⁻¹
- ¹⁰⁷⁸ Q_d Flow rate of diluate, m³ s⁻¹
- 1079 r_i Safety factor
- 1080 *R* Resistance, Ω
- ¹⁰⁸¹ Sh Sherwood Number
- 1082 *t* Time, s
- ¹⁰⁸³ *T* Temperature, K
- $t^{AEM,CEM}$ Transport numbers of the AEM and CEM
- 1085 membranes
- 1086 $t^{+,-}$ Minimum of the anion and cation transport num-
- 1087 bers

V_{el} Electrode potential, V	108
V Voltage, V	1089
V^{cell} Volume of a cell, m ³	1090
V^{tank} Volume of tank, m ³	109
W Stack width, m	109
z Ion charge	109
ϕ_A Open-area porosity of the spacer	1094
ϕ Current leakage factor	109
au Control time, s	109
Superscript and subscript	109
0 Position in the tank	109
AEM Anion exchange membrane	109
<i>CEM</i> Cation exchange membrane	110
<i>y</i> Position at the segment <i>y</i>	110
<i>Y</i> Position at the segment <i>Y</i>	110
τ_i Control time step <i>i</i>	110