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Generalized Disjunctive Programming Model for Optimization of Reverse Electrodialysis Process

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Abstract: Reverse electrodialysis (RED), an emerging electrochemical technology that uses ion-selective membranes to directly draw electricity out from salinity differences between two solutions, i.e., salinity gradient energy (SGE), has the potential to be a clean and steady renewable source to reach a sustainable water and energy supply portfolio. Although RED has made notable advances, full-scale RED progress demands more techno-economic and environmental assessments that consider full process design and operational decision space from module- to system-level. This work presents an optimization model formulated as a Generalized Disjunctive Programming (GDP) problem to define the cost-optimal RED process design for different deployment scenarios. We use a predictive model of the RED stack developed and validated in our research group to fully capture the behavior of the system. The problem addressed is to determine the RED plant's topology and the working conditions for a given design of each RED stack which renders the cost-optimal design for the defined problem and scenario. Our results show that, compared with simulation-based approaches, mathematical programming techniques are an efficient and systematic approach to provide decision-making support in early-stage applied research and to obtain design and operation guidelines for full-scale RED implementation in real scenarios.

Keywords: renewable energy, blue energy, process synthesis, Pyomo, GDPopt, global optimization

1. INTRODUCTION

The progressive shift from conventional to low-emissions decentralized renewable power sources with reduced water needs will be decisive in reaching a sustainable water and energy supply portfolio. Reverse electrodialysis (RED), an emerging electrochemical technology that uses ion-selective membranes to generate electricity out from the chemical energy released when two solutions of different salinities mix, i.e., salinity gradient energy (SGE), has the potential of being a clean and steady renewable source to power the water sector from the embedded energy of waste streams.

Even though past research has validated RED-based electricity from waste streams as desalination concentrates or treated wastewater effluents (Gómez-Coma *et al.*, 2020), full-scale RED progress demands more techno-economic and environmental assessments that consider full process design and operational decision space from stack to the whole system. Our research group is working on a modelling tool to provide decision-making support for early-stage applied research and to extract design and operation guidelines for full-scale RED implementation in real scenarios.

We have developed and validated a predictive model of the RED stack to determine the most relevant working conditions and design parameters affecting RED performance (Ortiz-Imedio *et al.*, 2019; Ortiz-Martínez *et al.*, 2020; Tristán, M. Fallanza, *et al.*, 2020); This rigorous model allow us to assess the retrofit of medium-to-large-sized seawater reverse osmosis desalination plants across the globe with a RED-based energy recovery system through simulation (Tristán, Marcos Fallanza, *et al.*, 2020). If all SGE were harnessed, RED could meet ~40% of the desalination plant's energy demand almost in all locations. However, energy conversion losses and untapped SGE decline it to ~10%. These results show that there is a gap to bridge between the thermodynamic limit and the actual energy the RED system produces. Several variables affect the RED process, adding complexity to define optimal system designs by conventional trial and error evaluations. Hence, we propose an optimization model formulated as a Generalized Disjunctive Programming (GDP) problem to define the cost-optimal RED process design for different deployment scenarios.

2. OPTIMIZATION MODEL

2.1 Problem statement and superstructure definition

The formulation of the RED process synthesis problem can be stated as follows. Given the site-specific working conditions (i.e., the high-saline, HC, and low-saline, LC, feed streams' concentration, total flow rate, and temperature) and the stack design of all RED units (number of cell pairs, membrane's properties, spacers' thickness), the problem is to determine the RED plant topology (i.e., number and hydraulic arrangement of the RED units) and the working conditions (i.e., the HC and LC concentration and flow rate, electric current) of each RED

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stack in the RED plant that maximizes the net power output and net energy yield from the feed streams while minimizing cost of the RED process for the defined problem and scenario.

The superstructure of alternatives follows the Pyosyn Graph (PSG) representation (Chen, Liu, *et al.*, 2021). The RED process PSG representation in Fig. 1 consists of the following representation elements:

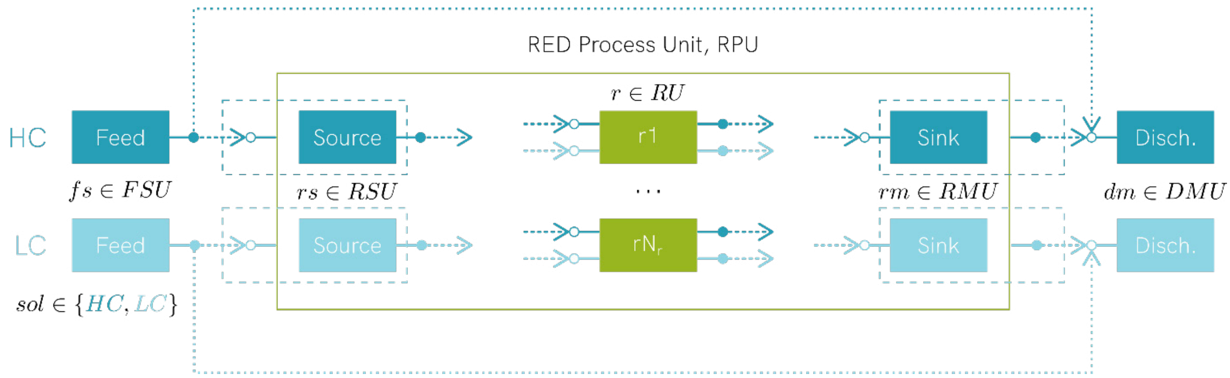

Figure 1 Superstructure representation of the RED process.

(a) The RED Process Unit (RPU), where discrete decisions on the selection of the RED units are made, that embeds: (a.i) the set of N_r candidate RED units $r \in RU = \{r1,\ldots,rN_r\}$; the additional set of (a.ii) source $rs \in RSU$ and (a.iii) sink $rm \in RMU$ units for the high-salinity and low-salinity streams, i.e., $sol \in SOL = \{HC, LC\}$, that govern the material in- and outflows for the overall flowsheet, respectively, and specifications on feed or effluent qualities of the RPU.

(b) The set of concentrate and diluate feed units, $fs \in FSU$, and discharge units, $dm \in DMU$.

(c) The inlet and outlet ports $p \in P = P_{out} \cup P_{in}$, —or mixers and splitters, respectively—, where flows of material at the unit interface with other process units may take place.

(d) The set of streams or feasible pairings of outlet ports to inlet ports, $s \in S \subseteq P_{out} \times P_{in}$, defined considering the following screening rules:

(d.i) The feed units, *FSU*, supply the concentrate and diluate feed streams to the RED Process Unit (RPU), while the discharge units *DMU* collect the exhausted high- and low-concentration RPU effluents, and the unused feed streams from the feed units *FSU*.

(d.ii) Within the RPU, the auxiliary source units, *RMU*, supply the concentrate and diluate streams coming from the feed units *FSU* to one or more of the active RED units. Once the active *RU* exploits SGE from the inlet streams, the exhausted effluents may be recycled back, sent to other active *RU* for reuse, or may be directed to the sink units, *RMU*. The RPU effluent from *RMU* is disposed of in the overall discharge unit *DMU*.

(d.iii) No flow between the *RSU* and *RMU* is allowed, it can only take place between *FSU* and *DMU*.

(d.iv) Mixing between the concentrate and diluate streams only take place within the candidate RED units owing to the flow of ions from high-saline compartments to low-saline ones through ion-exchange membranes (IEMs).

2.2 Generalized Disjunctive Programming (GDP) model

We formulate the superstructure of alternatives as a Generalized Disjunctive Programming (GDP) problem coded in Python using the Pyomo algebraic modeling language (Hart *et al.*, 2017). The optimization model, based on the superstructure in Fig. 1, is shown by the set of equations (1).

$$
\begin{aligned}
\min obj = \;& f(x) \\
s.t. \;& g(x) \le 0 \\
& \begin{bmatrix} Y_r \\ r_r(x) \le 0 \end{bmatrix} \veebar \begin{bmatrix} \neg Y_r \\ B^r x = 0 \end{bmatrix} \forall\, r \in RU \\
& \Omega(Y_r) = True \\
& x \in X \subseteq R^n \\
& Y_r = \{True, False\} \,\forall\, r \in RU
\end{aligned}
\qquad (1)
$$

The objective function $f(x)$ minimizes the Levelized Cost of Energy (LCOE) of the RED process subject to inequality constraints (e.g., from process specifications) and equality constraints (e.g. from material, energy balances and thermodynamic relationships).

Variables x describe continuous decisions (e.g. molar concentrations, volumetric flows) of all feasible streams and the electric current of the candidate RED units.

The global constraints, $g(x) \le 0$, are equalities and inequalities describing specifications and physical relationships that apply for all feasible configurations in the superstructure, e.g., flow and mass balances of the feed, source, sink, and discharge units' inlet and outlet ports, and the upper and lower bounds on streams' variables (concentration and flowrate).

The disjunctions, corresponding to logical-XOR relationships, such that at most one disjunct in each disjunction is True, describe the existence or absence of the RED units within the RED process unit. Boolean variables Y_r indicates whether a given RED unit exists or not. If a unit exists (Y_r = True), the constraints $r_r(x) \le 0$ enforce the relevant mass and energy balances, thermodynamics, kinetics, or other physical/chemical phenomena taking place within the RED unit. When the unit is absent, the negation ($\neg Y_r$) sets to zero a subset of the continuous variables, and cost terms in the objective function through the $B^r\ x = 0$ constraints.

Mixing and splitting calculations when the port exists are included within the constraints $r_r(x) \leq 0$, and port absence in the linear constraints $B^r\ x = 0$. We adopt the no-flow approach for modeling an absent unit, enforcing that if a stream does not exist, no flow may take place between the corresponding outlet-inlet port pair.

When the RED unit is active (Y_r = True), the boundary conditions specify the concentration and flow rate of the streams entering and leaving the RED unit, linking the inlet port ri with the RED unit's inlet compartments (i.e., $x = 0$), and the outlet from the set of cell pairs (i.e., $x = L$) with the outlet port ro of the RED unit. When the RED unit is absent ($\neg Y_r$), we enforce connected streams' null flowrate and minimum concentration, and the net power output and cost terms in the objective function are set to zero.

Other types of logical relationships ($\Omega(Y_r) = True$) are described using logical propositions.

2.3 Flow and mass balance formulation

We formulated flow and mass balance equations considering total flows (volumetric flow rate, Q in m^3/h) and species composition (molar concentration of sodium chloride, C in mol/m^3), of the high- and low-saline streams.

The mixer's balances apply to the inlet ports of the discharge units, the sink units, and the active RED units (i.e., when Y_r = True). Mixing equations are nonlinear and nonconvex due to the bilinear terms from the product of volumetric flowrate times molar concentration, which challenges finding a global optimum.

The splitter's balances apply to the outlet ports of the feed units, the source units, and the active RED units (i.e., when Y_r = True). Splitting equations are linear and convex.

2.4 Bounds on variables

Each RED unit has upper limits on the flowrate, according to the maximum linear cross-flow velocity along the channel's length of the RED stack as per the manufacturer's specifications. The lower bound on velocity and flowrate in the low salinity channels is a designer's choice.

The upper bound on concentrate streams' concentration could be as high as the maximum concentration of the feed streams, *in* (if there are multiple feed alternatives), while for the diluate streams, the molar concentration could be as high as the concentration reached after the complete mixing of the concentrate and diluate stream (if reached thermodynamic equilibrium). The opposite holds for the lower bound on the concentration of the concentrate and diluate streams.

2.5 RED stack model formulation

We used a simplified version of the RED stack's rigorous model from our research group (Tristán, M. Fallanza, *et al.*, 2020), to find a middle-ground between model fidelity and tractability.

The semi-rigorous model is a system of differential and algebraic equations defining RED's performance from cell pair to module scale. A cell pair is a repeating unit stacked in series to form the RED pile, assembled by alternating a cation- and an anion-exchange membrane (CEM and AEM, respectively) with two adjacent spacer-filled compartments where the concentrate and the diluate water streams flow.

The cell pair model describes the main transport mechanisms across membranes, mass balances, distributed pressure drop, and the electric variables, i.e., the potential difference and internal resistance, within a cell pair.

At RED stack's scale, the model computes the overall values (electric potential, electric current, internal resistance, pressure drop) of the whole set of series cell pairs averaging each distributed variable in the flow direction (taking the integral over the length domain, x). The model also computes the gross and net power output (deducting the pumps' power consumption from the gross power output) to define performance metrics such as power density, energy yield, and specific energy of the RED stack. The net power of the RED unit is also used to compute the net power output of the system.

The reader is referred to work (Tristán, M. Fallanza, *et al.*, 2020) for more details on the RED stack model.

As nonlinear optimization solvers are unable to handle integrals or differential equations directly, we apply the backward finite difference method and the trapezoid rule to reformulate first-order ordinary differential equations, and integrals into algebraic equations, respectively.

2.6 Assumptions

We assumed:

(a) The feed streams are pure sodium chloride, ideal aqueous solutions (i.e., activity coefficients equal to 1), thus neglecting the non-idealities of aqueous solution and existence of other species that would undermine the RED performance.

(b) There is no non-ohmic contribution in the internal losses ascribed to concentration polarization phenomena in the concentrate and diluate membrane-solution interfaces and owing to concentration gradient decline along the main flow direction. We only consider ohmic contribution due to solutions' ionic conductivity and membranes' ionic resistance.

(b) Membranes' permselectivity and ionic resistance are constant regardless of solutions' concentration and temperature.

(c) There is no water transport due to osmosis from the low-saline side to the high-saline one across membranes.

(d) Salt diffusivities in the membrane phase are constant independent of the concentration and temperature.

(e) All cell pairs behave equally, as we assumed no fluid leakage or ionic shortcut currents in the RED stack's manifolds.

(f) Co-current flow of the high- and low-concentration streams.

(g) The RED system operates under isothermal and isobaric conditions.

2.7 Objective function: Levelized Cost of Energy (LCOE)

The objective of the GDP problem is to minimize the LCOE of the RED process. The LCOE (USD_{2019}/kWh), a common metric to benchmark different renewable power technologies, estimates the average cost per unit of energy generated across the lifetime of a power plant. The LCOE considers operating (OPEX in USD_{2019}/year), and capital costs (CAPEX in USD_{2019}) annualised over the expected lifetime of the plant, LT in years, using the capital recovery factor (CRF) given in (15) with an interest rate r, that altogether define the total annual cost of the RED system.

Assuming the energy provided annually is constant during the lifetime of the project, the $LCOE$ reduces to (14). The annual energy yield (kWh/year) of the RED plant working at full capacity, i.e., 8760 full load hours per year, is corrected with a load factor, LF, of 90% (i.e., RED works 8000 hours each year). The summation of the net power output over the candidate RED units yields the nominal capacity of the RED system (16), i.e., the total net power output, TNP, in kW.

$$LCOE = \frac{CRF\,CAPEX + OPEX}{TNP\,8760\,LF} \tag{14}$$

$$CRF = \frac{r}{1 - (1 + r)^{-LT}} \tag{15}$$

$$TNP = \sum_{r \in RU} NP_r \tag{16}$$

To estimate the capital investment, we determine the cost of RED units, pumps, and civil and electrical infrastructure cost.

$$CAPEX = CC_{stack} + CC_{pump} + CC_{civil} \tag{17}$$

The annual operating cost comprises the electricity cost from pumps' consumption, the replacement cost of the RED membranes, and maintenance and labor costs (as 20% of CAPEX).

$$OPEX = OC_{pump} + OC_{IEMsrep} + 0.2\,CAPEX \tag{18}$$

Wherever needed, all currencies were converted to USD_{2019} according to historical average exchange rate of the corresponding publication year.

2.8 Solution strategy

For the solution of the non-convex GDP problem, we use the logic-based solver GDPopt built on Pyomo.GDP, an open-source ecosystem for GDP modeling and development, built on top of the Pyomo algebraic modeling language (Chen, Johnson, *et al.*, 2021).

We apply the Global Logic-based Outer Approximation (GLOA) algorithm (Lee y Grossmann, 2001; Chen *et al.*, 2018) to solve the GDP problem. This strategy decomposes the GDP into reduced NLP subproblems and Master MILP problems, to avoid "zero-flow" numerical issues arising in nonlinear design problems when units or streams disappear.

The GAMS/CPLEX MILP subsolver and GAMS/MSNLP(IPOPTH) NLP subsolver were used through Pyomo-GAMS interface using GAMS 34.1.0.

3. CASE STUDY: ENERGY RECOVERY FROM DESALINATION CONCENTRATE

We consider as a case study the retrofit of several medium-to-large capacity seawater reverse osmosis (SWRO) desalination plants distributed worldwide with a RED-based energy recovery system. The RED plant retrieves electric power from desalination's concentrate effluent reversibly mixed with different available low-salinity sources (e.g., WWTP effluent, seawater, river water). Data on concentration, temperature, and volumetric flow rate of the high- and low-saline feed streams are taken from (Tristán, Marcos Fallanza, *et al.*, 2020).

We compare the performance of the fixed series-parallel arrangement of the RED units, set in our previous assessment (Tristán, Marcos Fallanza, *et al.*, 2020), with the hydraulic topology the optimization model predicts in two pricing scenarios: assuming the current price of membranes (i.e, \$98 per membrane area as the manufacturer quoted) and the price that break-evens the RED system cost (i.e., \$1.12 per membrane area).

The study restricts to one parallel branch setting the flow rate and concentration of the inlet streams to each parallel branch equal to the optimal working conditions of the 1st RED unit in series for two hydraulic arrangements:

(a) Series layout, from our former study, imposing that the outlet stream of the RED unit is fed to the inlet port of the following series unit, so recycling or alternative reuse options of the outlet streams are not allowed.

(b) GDP layout, leaving the interconnection between the superstructure units free as a discrete decision.

The upper limit of parallel branches was set considering that the RED system treats desalination's concentrate effluent in full, assuming no shortage of low-saline feed stream. Then, the net power output of the RED system scales with the number of parallel branches.

The assessment refers to a commercial RED unit with an assumed number of cell pairs representative of industrial-scale stacks. Data on RED stack's parameters and process specifications can be found in (Tristán, Marcos Fallanza, *et al.*, 2020).

4. RESULTS AND DISCUSSION

The optimization model provides an optimized RED's flowsheet design in all scenarios improving the cost-competitiveness of the system compared with the series-parallel arrangement. As an illustrative example, we show the optimization results for the RED-retrofit of Barcelona-Llobregat SWRO desalination plant (Sanz y Miguel, 2013; Tristán, Marcos Fallanza, *et al.*, 2020).

The RED process problem with a fixed configuration has 8749 constraints (1897 nonlinear), 15 disjunctions, and 8329 variables (30 Boolean, 8299 continuous). When the RED units' arrangement is not fixed, the problem size increases to 9229 constraints (1927 nonlinear), and 8807 variables (8777 continuous) with the same number of disjunctions and Boolean variables.

The CPU time for solving these two problems (Table 3) on a machine running Windows 10 (x64) with 6 cores processor (Intel® Core™ i7-8700 CPU @3.2 GHz) and 16 GB of RAM was 471 s and 518 s, respectively.

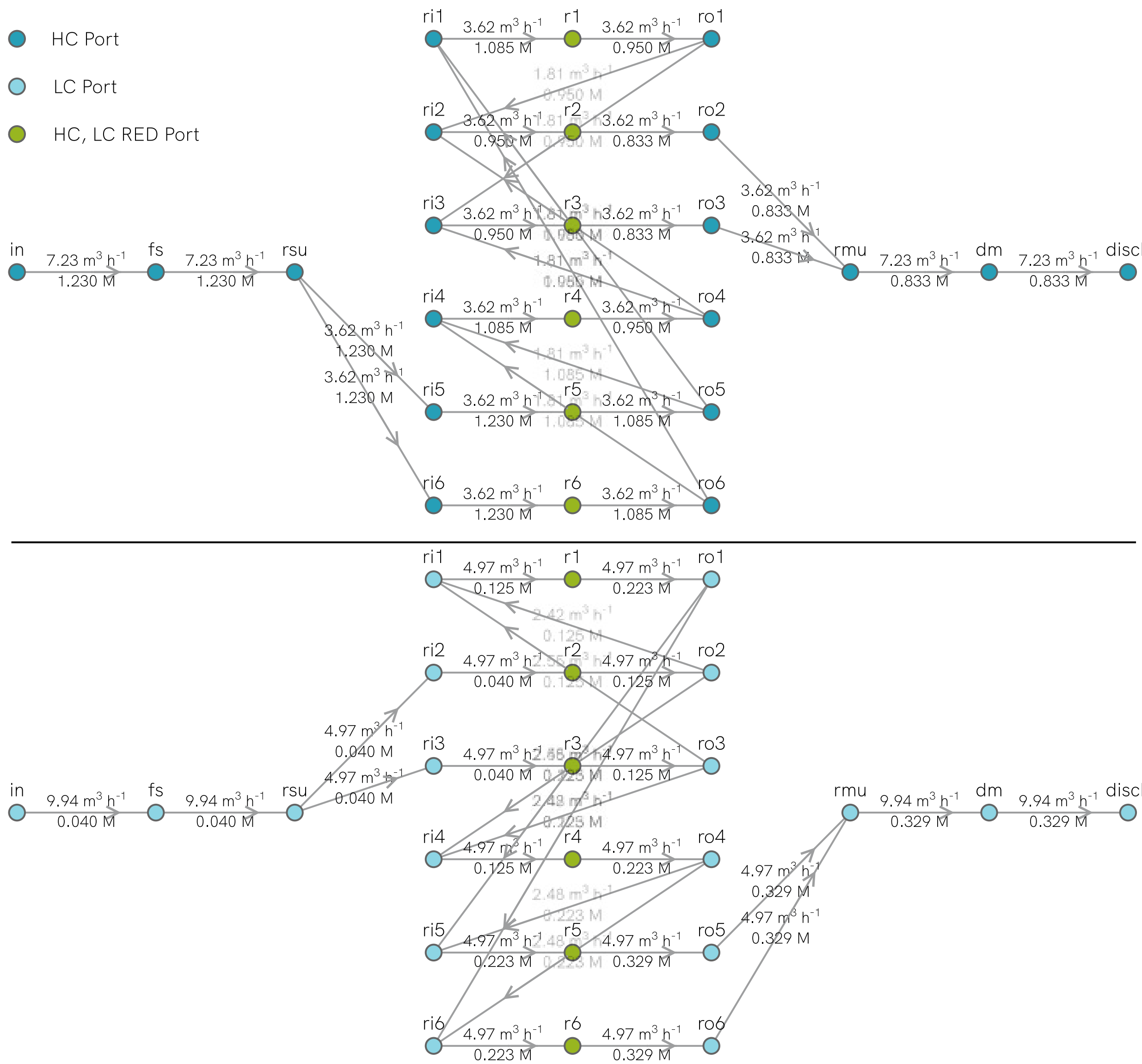

Figure 2 Port connection of a parallel branch in the RED process' optimal flowsheet design. Case study: N_r = 15; RED stack working conditions (HC/LC): 1.23/0.04 M, 7.24/9.87 m^3/h, 19 °C. GDP (decision) layout; IEMs price = \$1.12/m^2.

Table 1. Optimization results of a parallel branch in the RED system for the fixed and decision hydraulic arrangement of the RED units, and the current and optimistic membranes' price scenarios.

Layout	IEMs (\$/m^2)	#·RED untis	TNP (kW)	LCOE (\$/MWh)	CAPEX (k\$)	OPEX (k\$/year)
Series (fixed)	1.12	5	2.99	162	21.32	1.73
	98	1	0.95	369	27.77	2.09
GDP (decision)	1.12	6	3.45	147	22.04	1.83
	98	1	0.95	369	27.77	2.09

Case study: N_r = 15; RED stack working conditions (HC/LC): 1.23/0.04 M, 7.23/9.94 m^3/h, 19 °C

The GDP optimization model renders a flowsheet design (Fig. 2) that reduces the LCOE by 9% and increases the net power output of the RED system by 15%, which in turn could save around 16% of the desalination plant's specific energy consumption sourced by the local grid mix if economies of scale drive two orders of magnitude down the current cost of membranes.

5. CONCLUSIONS

In this work, we propose a GDP optimization model to systematically synthesize and optimize the RED process for electricity production from salinity gradient energy. The goal is to define the hydraulic topology, that is, the number and hydraulic layout of the set of candidate RED units, and the working conditions of each RED stack that minimize the LCOE, used as a metric to measure RED process economic competitiveness in a given application scenario. Simulation-based approaches on conceptual design fail to handle complex systems with many degrees of freedom, which may lead to suboptimal solutions. Hence, to illustrate how mathematical programming techniques can enhance RED process conceptual design over conventional trial-and-error procedures, we evaluated RED energy recovery from desalination's concentrate effluent around the world for two hydraulic configurations: (i) a fixed series-parallel arrangement of the RED units with no recycle or additional reuse alternatives of the exhausted water streams after SGE retrieval, and (ii) freeing the layout to accommodate all feasible reuse and recycle alternatives. Compared with the simulation-based approach from our previous study, the optimization model provides a RED's flowsheet design that improves the cost-competitiveness of the system in all scenarios. Our results reveal mathematical programming techniques as an efficient and systematic decision-making approach over simulation alone to advance RED's Technology Readiness Level. The process synthesis model can be a valuable tool to assist RED field demonstration and deployment stages in real environments.

Future work will test different solution strategies and problem formulations to improve the computational effort and robustness of the model. It will also extend the superstructure of alternatives with more discrete and continuous decision variables, related to RED stack design (e.g. the number of cell pairs, membranes' and spacer's design) and the RED system (e.g., adding auxiliary equipment as DC-AC inverters, or processes as pre-treatment of feed's solutions), and consider environmental concerns through multi-objective optimization.

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