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# Critical Mineral Separations: Opportunities for Membrane Materials and Processes to Advance Sustainable Economies and Secure Supplies

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critical mineral separations, high-throughput experiments, solute-tailored selectivity, membrane-based processes optimization, machine learning, water–energy nexus

## Abstract

Sustainable energy solutions and electrification are driving increased demand for critical minerals. Unfortunately, current mineral processing techniques are resource intensive, use large quantities of hazardous chemicals, and occur at centralized facilities to realize economies of scale. These aspects of existing technologies are at odds with the sustainability goals driving increased demand for critical minerals. Here, we argue that the small footprint and modular nature of membrane technologies position them well to address declining concentrations in ores and brines, the variable feed concentrations encountered in recycling, and the environmental issues associated with current separation processes; thus, membrane technologies provide new sustainable pathways to strengthening resilient critical mineral supply chains. The success of creating circular economies hinges on overcoming diverse barriers across the molecular to infrastructure scales. As such, solving these challenges requires the convergence of research across disciplines rather than isolated innovations.

## INTRODUCTION

Metal and mineral resources are critical to electrification, green energy technologies, and components in high-performance computers. The United States has identified 50 critical minerals, including lithium, several transition metals (e.g., cobalt and nickel), and rare earth elements (REEs) (1). These resources are critical because reducing their supply would detrimentally impact decarbonization goals and economic prosperity. For instance, lithium and cobalt are vital components in rechargeable batteries, including those used in electric vehicles (EVs) (2). In 2021, ~45% and ~20% of worldwide lithium and cobalt production, respectively, were used to manufacture EV batteries (3). By 2030, lithium demand is expected to increase four- to sixfold, and the development of EV batteries will consume 70–80% of all lithium (3). REEs are critical components in powerful, high-flux magnets that replace gearing systems in EVs and harness energy more efficiently in wind turbines (4). Every megawatt of rated wind turbine capacity requires several hundred kilograms of REEs (5).

Given the importance of critical minerals, governments are investing billions to secure and reinforce domestic supply chains. For instance, the United States and the European Union recently passed laws promoting the domestic manufacturing of semiconductors and their associated equipment and raw materials [Chips and Science Act, H.R. 4346, 117th Congr. (2022)]. Securing domestic supply chains will require countries to identify new critical mineral sources capable of meeting projected demands and refine current processes to conform to increasingly stringent environmental and human rights regulations (6, 7). The small footprint and customizable nature of membrane modules provide opportunities to treat materials closer to their point of origin, purify dilute feed streams, and replace harmful solvents and precipitating agents used in traditional separation techniques.

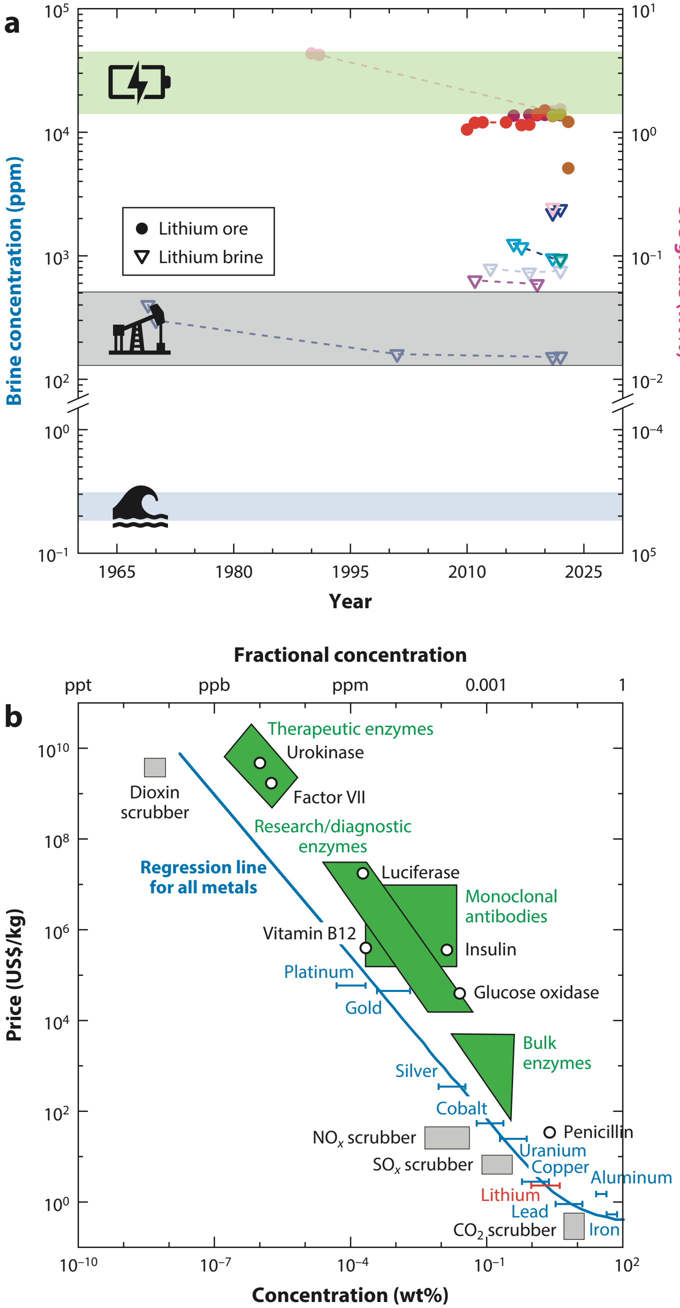
## CHALLENGES IN SOURCING CRITICAL MINERALS MAY SLOW THE ADOPTION OF GREEN TECHNOLOGIES

While the demand and need for critical resources continue to grow, their availability in Earth's crust physically constrains the supply of primary sources. Resources with the highest grade are consumed first, leaving behind ores with reduced concentrations. For example, the concentration of copper in ores mined within the United States has decreased from 1.8% to 0.40% over the past 100 years (8). The decreasing trend in ore grade is consistent with reports for other minerals, and similar trends are emerging for critical resources (9). **Figure 1*a*** presents the concentration of lithium mined from ores (circular data points) and salar brines (triangular data points) with respect to time [see the **Supplemental Material** for tabulated data, including the recently reported McDermitt caldera reserves (10)]. Several mines have data spanning over 20 years, which begin to reveal long-term trends. For instance, the lithium brine concentration of the US Clayton Valley mine decreased from 400 mg $L^{-1}$ to 160 mg $L^{-1}$ from 1966 to 1988 (11). A similar decrease in ore grade, from 4.33% to 1.53% between 1990 and 2022, was recorded for the Australian Greenbushes mine (12, 13). The decrease in ore grade leads to more significant costs (e.g., increased transportation, water use, energy consumption, and greenhouse gas emissions) associated with extracting the mineral (14). This relationship is consistent with trends observed in the Sherwood plot for industrial gases, pharmaceutical products, and metals (**Figure 1*b***). Over 12 orders of magnitude, a

Supplemental Material >

power law dependence exists between the concentration of the product in the initial feed and the processing cost, implying that targeting more concentrated feed streams first is more cost effective.

Along with the continued exploitation of geological reserves, there exist opportunities to use unconventional mineral sources (e.g., brines from petroleum waste streams and e-waste recycling).



*(Caption appears on following page)*

**Figure 1** (*Figure appears on preceding page*)

Lithium concentration in select sources. (*a*) The lithium concentration found within ores and brines is plotted with respect to time. The data are obtained from annual reports of mining companies and tabulated within the **Supplemental Material**. Circles and triangles represent the lithium concentration of ores and brines, respectively. Data points of the same color correspond to the same mine. The dotted lines are meant to guide the eye. The green-, gray-, and blue-shaded regions correspond to the concentration of lithium in electric vehicle battery packs (∼1–3 wt%) (2), oil field brines (∼100–300 ppm) (15), and seawater (∼0.17–0.25 ppm), respectively. (*b*) A Sherwood plot presents the price to process a material versus its concentration in the initial feed stream (16, 17). Industrial gasses are indicated in gray (18), pharmaceutical products in green (16), and metals in blue (19). Panel *b* replotted with permission from Reference 17. Abbreviations: ppb, parts per billion; ppm, parts per million; ppt, parts per trillion.

**Supplemental Material >**

This opportunity is motivated by the fact that, because of declining ore grades, the concentration of critical minerals in consumer products is starting to approach, and in some cases surpass, the concentration of minerals within ores and brines. For instance, the average concentration of lithium within EV batteries (**Figure 1**, *green stripe*) (2) is greater than that in salar brines and comparable to that in solid ore reserves (15). Likewise, the lithium concentration within oil field brines (**Figure 1**, *gray stripe*) is comparable to that of salar brines (15). Using secondary sources could significantly offset demand and extend the life of natural reserves. As one example, it is estimated that 1.3 TWh of batteries will reach the end of their life in 2040. If recycled, the spent battery waste that will be generated will account for 12% and 5% of the total cobalt and lithium demand, respectively (20).

Although secondary sources are a viable supplement, several challenges remain. The first technical issue is the variability associated with the composition of the recycling stream. Specifically, product development has resulted in materials possessing many elements, some of which are challenging to isolate from one another (8). For instance, whereas high-performance circuits used only 12 elements in the 1980s, today, their manufacture regularly applies more than 60 (21). Although the components of circuits are well known, recycling is complicated by the current infrastructure, whereby electronic waste is combined, crushed, and leached in large batches. Therefore, the elemental composition of the leachate varies from batch to batch. Advances in processes that can be reoptimized rapidly to perform the desired separation are necessary, but this variability will not be overcome with technical advances alone. As such, there exists a need to implement infrastructure and machinery that can systematically collect, sort, and disassemble products to provide homogeneous recycle streams (22). Successful recycling programs for goods regulated by law (e.g., lead acid batteries and catalytic converters) demonstrate the potential for such infrastructure (23, 24).

In this review, we argue that advanced membrane technologies are well-positioned to address declining feed concentrations in ores and brines, the variable feed concentrations encountered in recycling, and the environmental issues associated with current processing methods, thus providing new, economically viable pathways to strengthen critical mineral supply chains. We begin by highlighting the significant energy and resource demands associated with current critical mineral recovery and purification processes. We then enumerate opportunities for process intensification offered by membrane separations. Subsequently, we describe two challenges hindering these opportunities. First, we highlight the need for better physical insights and quantitative descriptors of how membranes function in complex, multicomponent mixtures. Next, we focus on how membranes that leverage molecular recognition can be engineered to empower solute-selective separations. We conclude by detailing how digital engineering can accelerate membrane technology development and deployment by facilitating research convergence across disciplines.

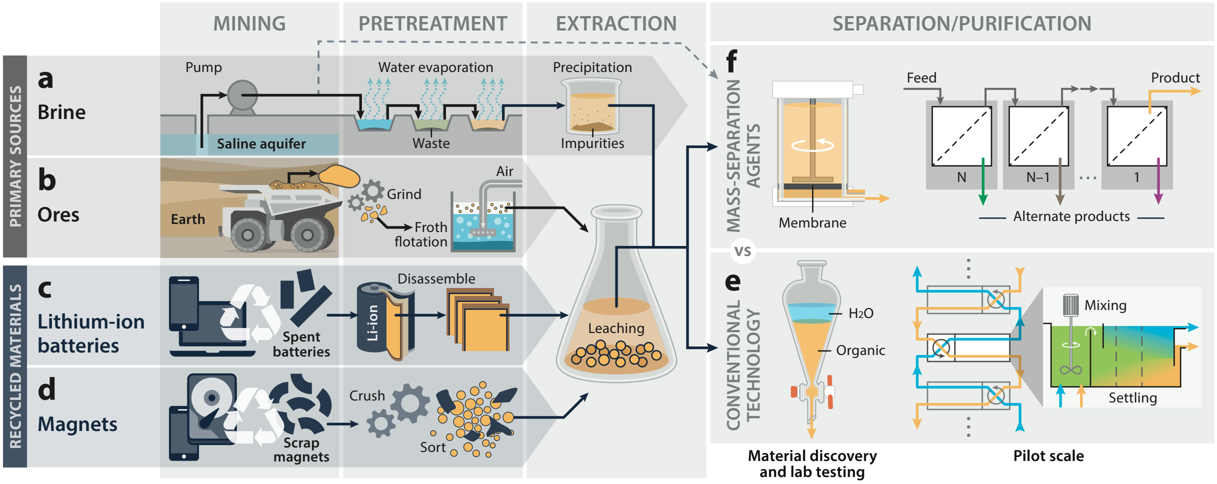


**Figure 2**

Independent of source, critical materials possess similar processing routes. (*a*) Conventionally, brines are pumped into a series of large evaporating ponds to increase lithium concentration. After removing competing ions, lithium is precipitated out of the solution as lithium carbonate. Depending on the desired purity, additional purification steps may be required. (*b*) Lithium from ores follows a similar path in which beneficiation (i.e., pretreatment) increases the lithium concentration. The mineral is subsequently leached into the solution before undergoing a series of purification steps. (*c*,*d*) Urban mining involves collecting consumer electronics (e.g., lithium-ion batteries and magnets). Product disassembly, crushing, and sorting occur before hydrometallurgical or pyrometallurgical processes harvest the critical materials. (*e*) Traditional liquid–liquid extraction and ion exchange processes separate critical materials from complex milieus. (*f*) Membranes that can fractionate solutes into multiple product streams can treat salar brines directly. Likewise, membranes capable of separating charged ions from one another can replace many of the harsh chemicals used to purify ores and recycled products.

## CRITICAL MINERAL PROCESSING TECHNOLOGIES REDUCE THE SUSTAINABILITY IMPACT OF THE GREEN ENERGY TECHNOLOGIES THEY INSPIRE

The extraction and purification of critical minerals have changed little over the past century, leaving in place processes that are inefficient and environmentally taxing. The Salt Lake brines in Chile, estimated to constitute 35.7% of global lithium reserves, are one example (**Figure 2*a***) (25, 26). The brines, located in regions with high temperatures and little rainfall, are pumped into a series of large evaporating ponds. As water evaporates, the concentration of salts increases. Competing ions (e.g., potassium, sodium, and magnesium) precipitate, simultaneously purifying and enriching the concentration of lithium (27, 28). After reaching a predetermined concentration (typically 6% lithium chloride), lime (i.e., calcium hydroxide) and sodium carbonate are added to remove magnesium and calcium. Boron, present as boric acid/borate ions, is a contaminant in specific brines (e.g., the Salar de Atacama) that is removed using a series of liquid–liquid extraction (LLE) stages (11). Subsequently, the addition of sodium carbonate, followed by filtration of the precipitate, produces lithium carbonate (at a purity >90%) (29). Additional purification steps may be required to achieve higher purities depending on the end-use application. The lime soda evaporation process is chemically expensive, consumes a large amount of water, and requires 12–18 months to produce lithium, hindering the ability of supply chains to adjust rapidly to economic demands. Membranes capable of fractionating brines into multiple product (e.g., lithium and potassium) and by-product (e.g., boron and magnesium) streams can remove the need for pretreatment and extraction steps. Additionally, membrane distillation or solar evaporators may enable brine processing in non-arid climates and concentrate streams before the final product is precipitated (30). Systematic comparison of conventional (e.g., hydrometallurgy) and membrane-based critical mineral processes, especially from a life cycle assessment perspective, is needed to assess the long-term viability of such opportunities. In other applications (e.g., postcombustion carbon capture), process-scale benefits of using membranes have been investigated, showing that membrane systems can have lower emissions (31, 32) and be more energy efficient (32, 33) and cost-effective (33) compared to traditional absorption- or solvent-based methods.

Membranes can improve the sustainability of ore processing by reducing the use of chemical reagents. After mining, the ore is reduced in size (e.g., through crushing and grinding) and separated based on physical (e.g., density and magnetic susceptibility) and chemical properties (8). The beneficiation steps remove undesired materials to increase the concentration of the desired metal. Subsequently, critical materials are extracted from the ore using a mix of hydrometallurgical (solution-based chemical reaction) and pyrometallurgical (high-temperature liberation) processes (34). The unit operations are highly specialized and require large amounts of energy and water. For example, for every tonne of $Li_2CO_3$ produced from ore, 7.3 tonnes of $Li_2O$ concentrate (∼32.8 tonnes of virgin ore), 1.71 tonnes of $H_2SO_4$, 2.05 tonnes of $Na_2CO_3$, 0.7 tonnes of $CaCO_3$, 40 tonnes of water, and 142,370 MJ of energy are consumed (35). This energy requirement is equivalent to powering the average US household for approximately 4 years (36). Based on the utility costs of these processes, it is more desirable to process material in an off-site, centralized location with available process utilities and economies of scale. For lithium, beneficiation methods aim to produce an approximately 6% (by weight) $Li_2O$ concentrate. Whereas beneficiation decreases the quantity of material that must be transported to centralized facilities, onsite leaching using sustainable, environmentally responsible solvents (37–40) can further decrease greenhouse gas emissions related to transportation. Specifically, depending on the source, energy and greenhouse gas emissions may be reduced up to 10% (35). At the centralized facility, the concentrate is heated to ∼1,000°C and baked at 250°C in sulfuric acid to leach the lithium into solution (11, 19, 41). Similar to the method used in brine treatment, calcium and magnesium impurities are removed by adding lime and sodium carbonate, respectively. The resulting solution is concentrated before precipitating lithium carbonate (∼95% purity). Although specific reagents (e.g., $H_2SO_4$) are required to leach lithium from the ore, the leachate can be purified with membrane technologies. In this regard, membranes that remove magnesium and calcium from the leachate could eliminate $CaCO_3$ and decrease $Na_2CO_3$ use by up to 26.5% (19, 35). By reducing reagents and waste, high-grade lithium reserves could be harnessed with sustainable processes that meet stringent health and environmental standards associated with increasing standards of living.

## MEMBRANES OFFER ADVANCES IN PROCESS INTENSIFICATION

Current ore-refining practices are ripe for process intensification, especially via emerging technologies such as membranes (42–44). REEs are mined and leached in analogous ways to the treatment of lithium ores; however, differences in purification practices emerge in LLE technologies (45–48). Whereas 4–6 LLE stages are needed to remove contaminants from lithium ores and brines, REE recovery processes may possess as many as 1,500 stages due to the small differences in charge and size of these elements (11, 47). As a result, the solvents, acids, and bases used in LLEs and the subsequent stripping steps generate large amounts of waste. Membranes with solute-selective transport mechanisms can replace LLE technologies and reduce the environmental impact of these processes (49, 50).

The flexibility associated with membrane-based systems enables improvements in process intensification with respect to feed stream variability. Conventional separation technologies (e.g., LLE) use large settling tanks that take hours to days to reach a steady state. In contrast, membranes possess a smaller footprint and hold-up volume, which, coupled with the ability to adjust pressure differentials rapidly, allows membrane separation technologies to be reoptimized subhourly (51). The ability to respond more quickly to disturbances is a significant advantage of membrane-based systems, especially considering possible daily feed variabilities from received shipments of diverse recycled materials. Moreover, the modular nature of membrane designs and evolving fit-for-purpose materials enables the rapid retrofitting necessary to meet the desired separation standards, which may evolve as new battery and electronics chemistries come to market (24). Additionally, modules enable staged separations, which can be optimized to reduce energy costs and chemical reagents even as membrane performance changes with time. Therefore, pushing toward tailored membrane technologies will enable practical, long-lasting separations solutions.

The modular design also enables system flexibility and customization, which have proven critical to the success of vapor-phase (e.g., distributed ammonia) and liquid-phase (e.g., decentralized water) membrane-based separations (52–55). We propose that distributed separations would enable a paradigm shift in critical mineral supply chains, a sharp reduction in intermediates transported, and more flexibility to respond to changing feed-side, recycling, and intermediate market conditions. For example, Chile's Salar de Atacama lake contains lithium and potassium concentrations four times larger than those found in China's Qaidam Lake (56). Therefore, modular membrane technologies can facilitate tailored separation configurations for site-specific constraints. Furthermore, the decreasing ore and brine quality over time introduces gradual changes in feed concentration. Thus, processes must be adaptable, as no one-size-fits-all solution exists in terms of either space or time. The success of staged membranes in other arenas (e.g., carbon dioxide capture and desalination) sets the stage nicely for this archetype that can leverage advances in modeling and optimizing staged separations to positively impact critical minerals recovery (49, 57–60).

Although membrane materials and unit operations for critical mineral separations have been refined over the past decade, these processes exist primarily on the laboratory scale and generally represent designs tailored toward a single feed stream. For example, technologies proposed to process brine from lakes in China feature electrodialysis (ED) with and without bipolar membranes and, in some instances, additional nanofiltration (NF) and reverse osmosis steps (61, 62). Although both pathways may be viable, exploring customization is expensive and time consuming, especially considering the increased complexity of a growing number of unique feedstocks containing critical minerals (e.g., diverse secondary sources from recycled consumer electronics). New modeling frameworks are needed to accelerate technology scale-up, from fundamental scientific advances to laboratory proof-of-concept, pilot validation, at-scale demonstration, and adoption, while recognizing that each critical mineral separation at a specific site is unique and requires customization that may challenge traditional economies of scale. These modeling frameworks require knowing how state-of-the-art membrane materials function and which properties to target for overall process improvement. Quantitative descriptions of the relationships between membrane performance and system operating conditions are needed to implement these modeling frameworks.

## MODELS CAPTURING MEMBRANE PERFORMANCE IN COMPLEX FEED STREAMS WILL GUIDE THE DESIGN AND IMPLEMENTATION OF FUTURE MATERIALS

Commercially available NF and ion exchange (IEX) membranes are already capable of distinguishing between monovalent and multivalent ions (61–64); as such, they could help to address separation opportunities related to the direct processing of brine, the removal of boron (65), and the replacement of chemicals used to precipitate contaminants in hard rock processing (66).

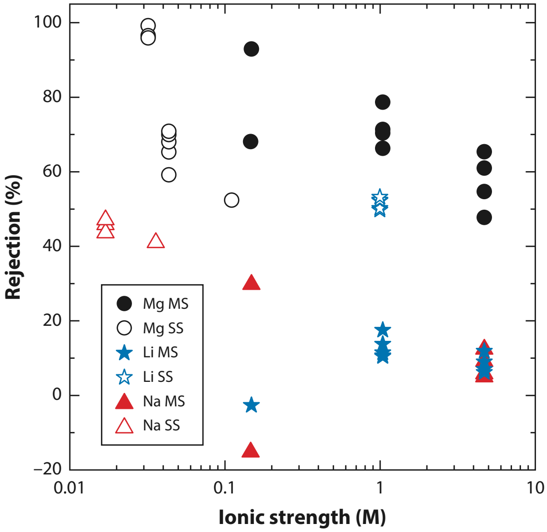


**Figure 3**

Nanofiltration performance in single- and mixed-salt systems (SS and MS, respectively). Solute rejection is plotted with respect to the ionic strength of the solution for single-component (*open symbols*) and multicomponent (*closed symbols*) systems. All data points correspond to nanofiltration membranes with similar charge densities and isoelectric points. The data are obtained from Foo et al. (56), Chu et al. (72), Ashraf et al. (73), Ouyang et al. (76), and Zhang et al. (77).

Electrostatic interactions between fixed charges on the membrane surface and ions dissolved in solution are essential to the function of NF and IEX membranes. Namely, the membrane repels coions (i.e., ions with the same charge as the fixed charge) and attracts counterions, consequently hindering or promoting ion partitioning, respectively (67). When an applied electrostatic potential drives transport, counterions permeate through the membrane preferentially. ED processes leverage this property by using stacks of alternating positively charged anion exchange and negatively charged cation exchange membranes to create devices capable of desalination and water softening (68, 69). Alternatively, when pressure-driven flow mediates transport, as is the case for NF processes, the reduced concentration of coions and the electroneutrality constraint lead to the retention of dissolved salts.

Due to the role that electrostatic interactions play in the function of NF membranes, multivalent coions are rejected more strongly than monovalent coions (**Figure 3**). Whereas the development of membranes for water treatment has often pursued high ion rejection, processes targeting the fractionation of ions will benefit from membranes that exhibit significant differences in the rejection of distinct ions. The concentration of dissolved ions and the concentration of fixed charge on the membrane influence the strength of the electrostatic interactions that promote ion retention (70). As a result, NF membrane performance depends on the ionic strength of the feed solution (71). For instance, $MgCl_2$ rejection in single-solute experiments decreases from ~95% to ~70% as the concentration increases from 17 mM to 1 M $MgCl_2$ (72, 73) (**Figure 3**). Concentration-dependent properties are also observed for IEX membranes (74, 75), which is important in the context of selective ED processes. Given the geographical and temporal variation in mineral streams referenced above, informed process design necessitates that these variations in NF and IEX membrane performance be quantified for relevant operating conditions.

Models describing the thermodynamics and transport phenomena for charged membranes and single-salt systems have been built through iterative feedback loops between experimentation and theory. Both top-down lumped parameter and bottom-up mechanistic models for describing ion transport have helped in designing and elucidating NF membrane materials and processes (78). Whereas initial models began by assuming ideal interactions within the membrane and solution phases (e.g., Donnan exclusion principles), emerging iterations include nonideal behavior [e.g., counterion condensation (79, 80), dielectric exclusion, and ion pairing (81–83)] in an attempt to expand the phase space over which the models are valid. For example, the Manning polyelectrolyte theory for counterion condensation to membrane materials has been applied to address experimental deviations in cation and anion sorption in their respective IEX membranes (79, 84). Similarly, physics-based models have captured ion-pairing effects of salts that do not fully dissociate (e.g., $Na_2SO_4$, $MgSO_4$) and can explain experimental observations related to elevated sulfate sorption into negatively charged membranes (81). Highlighting the interdependencies of these approaches, lumped parameter models, typically deployed for process-scale modeling, can be derived from mechanistic models. Insights from mechanistic models can also elucidate dependencies on fundamental material properties (e.g., pore size, pore size distribution, and surface charge).

The fundamental understanding extracted from mechanistic models provides valuable insights that empower the design of membranes with improved abilities to differentiate between ions of different valences (i.e., selectivity). For example, in single-salt systems, NF membranes perform best when the fixed charge concentration is greater than the concentration of ions in the bulk solution (70). For similar reasons, studies also suggest the need to increase the charge density in IEX membranes (85). To this end, many studies have engineered membrane surface chemistry [e.g., through the grafting of polymer brushes (77, 86), layer-by-layer schemes (87), or alteration of the interfacial polymerization process (88)] to increase the fixed charge density. Similar approaches have been used to modify IEX membranes (89). A prime example lies in layer-by-layer modified Nafion, in which films terminated with a positively charged polyelectrolyte hinder $Mg^{2+}$ permeation, leading to $Li^+$:$Mg^{2+}$ selectivities greater than 100 (89). In this context of controlling charge sign and density, numerous emerging materials platforms offer the potential for enhanced separation. For all of these potential approaches, to develop membrane processes capable of fractionating solutes, the charge density and material properties must be optimized with respect to process-scale design decisions. Although models can inform membrane and process design for solutions containing single salts, target applications (e.g., recycled waste or brines) comprise a complex milieu of ions that current theories do not accurately describe.

The targeted development of membranes for ion-selective processing technologies is hindered by a poor understanding of the complex interplay of phenomena that govern transport in multicomponent solutions. For example, consider the interesting behavior that can arise in binary salt solutions. In a single-salt (i.e., LiCl) solution, NF membranes reject $Li^+$ at ~50% (**Figure 3**, 1 *M* lithium solution). However, in a binary salt (i.e., LiCl + $MgCl_2$) system, $Li^+$ rejection decreases to 15% (**Figure 3**, 1 *M* multi-salt) (73). The decrease in salt retention observed in multicomponent systems arises due to ion–membrane and ion–ion interactions that increase partitioning and electromigration (56, 78). In the context of a monovalent and multivalent counterion sharing a coion, both the identity of the counterions and the membrane fixed charge concentration, which can be affected by solution pH, influence rejection (56). Models have succeeded in describing the transport of binary salt systems in which one of the components is present at trace concentrations. Nevertheless, opportunities exist to identify equivalent models for more complex systems (90, 91). To this end, fractionating industrially relevant feed streams through the selection of current (or design of future) membrane materials requires fundamental models or data-driven approaches capable of capturing the effects of complex multicomponent transport phenomena. Developing these models requires robust data over the broad phase space these systems may encounter.

## ENGINEERING MEMBRANE–SOLUTE INTERACTIONS CAN PRODUCE SOLUTE-SELECTIVE MEMBRANES

Significant opportunities exist for membranes that distinguish between ions of similar size and charge (e.g., REEs and transition metals). Currently, the principles of molecular recognition are applied to accomplish solute-selective separations in LLE, adsorption, and precipitation-based processes (50). Molecular recognition refers to using noncovalent (e.g., metal coordination and cation-$\pi$) interactions and complementary features (e.g., cavity size and shape) between a ligand and target solute to enable highly selective binding. In turn, this selective binding can be exploited to empower separations. For example, molecular recognition elements have been amended with alkyl tails to create extractants for use in LLE. The lipophilic alkyl tails sequester the extractants in the organic phase, thereby promoting the selective partitioning of target ions from the aqueous to the organic phase through selective association with the molecular recognition element. The molecular recognition elements within these extractants can be tailored to discriminate between ions based on small differences in molecular characteristics. For instance, two phthaloyl diamide ligands with a one-carbon length difference in cavity size showed distinct preferences between light (atomic numbers 57–64) and heavy (atomic numbers 65–71) REE uptake. The ligand with the smaller cavity size preferred complexation with the smaller, heavy REEs, whereas the light REEs favored the larger cavity (92). In another example, light and heavy REEs were separated from each other due to the number of oxygen atoms in the extractant (93). Despite the success of molecular recognition in these examples, LLE processes use copious amounts of organic solvent and struggle to address dilute feed streams in the 100–1,000 mg $L^{-1}$ range due to solvent loss between sequential separation cycles (94). Therefore, based on their process-scale advantages, there is interest in introducing molecular recognition elements within membrane architectures to enable transport mechanisms that exhibit solute-tailored selectivity.

For approaches that leverage molecular recognition to effect a separation, the binding strength between solute and ligand, which can be related directly to the equilibrium binding coefficient, $K$, is crucial to determining separation efficiency. Higher values of $K$, due to stronger binding interactions, allow for the treatment of more dilute feed streams. However, it is often more difficult to recover the target solute from the bound state when $K$ is large. Thus, when choosing the desired binding element, the optimal separation often will occur when $K^{-1}$ is approximately equal to the feed concentration as this choice balances energetic considerations related to solute binding and release. The extraction of neodymium from a mixture of other light REEs using di-(2-ethyl hexyl) phosphoric acid (DEHPA) as the extractant provides an example of the relationship between separation selectivity and concentration (95). At low DEHPA concentrations, negligible extraction of any REE ion was observed. At high DEHPA concentrations, all the REE ions were extracted indiscriminately at 99%. Both situations result in minimal selectivity. However, at intermediate extractant concentrations, the extraction of neodymium was preferred, resulting in the separation factor reaching values of 305 (95). A similar trend emerges for membrane architectures that attempt to recover target solutes through molecular recognition (**Figure 4**). Because they rely primarily on electrostatic interactions to promote separation, NF membranes have low binding strength and cannot distinguish between ions of the same valence. Materials that exhibit strong binding interactions can achieve solute-specific separations but do so through adsorptive rather than permeation-based mechanisms. At intermediate binding strengths, solute–ligand

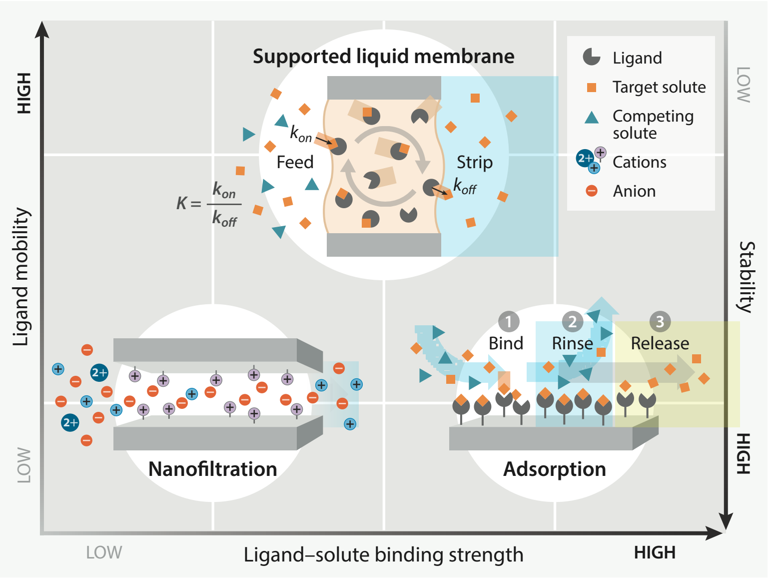


**Figure 4**

Comparison of relative ligand mobility and strength. Three major types of membrane transport mechanisms are shown in relation to their ligand mobility, stability, and strength of ligand–solute interaction. Nanofiltration membranes operate through a combination of steric hindrance and electrostatic interactions between the membrane and ions in bulk solution. The dynamics of the association and dissociation reactions between ligand and solute are integral to the properties of supported liquid membranes and adsorptive membranes. Polymer inclusion membranes and liquid-emulsion membranes are categorized under supported liquid membranes.

interactions can lead to effects [e.g., in supported liquid membranes (SLMs)] that promote the selective permeation of a target solute and ultimately achieve effective separations.

SLMs can achieve high selectivity while reducing organic solvent usage relative to LLE. As illustrated in **Figure 4**, SLMs are made by first dissolving an extractant in a water-immiscible organic solvent and then imbibing the resulting solution into the pores of a membrane. The SLM is placed between an aqueous metal-donating feed solution and a metal-receiving stripping solution. The molecular recognition element within the SLM promotes the selective transport of the target solute from the feed to the stripping solution. In a recent example of the selective separations that can be realized with this membrane architecture, an SLM was made by suspending a solution of tetraoctyl diglycol amide dissolved in a mixture of tributyl phosphate and Isopar L in the pores of microporous polypropylene. Specifically, the membrane recovered neodymium and dysprosium from NdFeB permanent magnets. After leaching the NdFeB magnets with 6 M $HNO_3$, the SLM extracted 99% of neodymium and dysprosium from the leachate, whereas only 10% of iron and boron were recovered (96). In this and other SLM examples demonstrating high REE recovery, the volume of organic solution within the pores of an SLM is orders of magnitude smaller than the volume required in LLE mixing tanks, thus reducing organic solvent demand dramatically while maintaining separation efficiency (97).

Experimental and theoretical studies have established a robust understanding of the mechanism underlying transport through SLMs. Namely, metal extraction occurs in three steps (**Figure 4**): (*a*) The carrier (i.e., the dissolved extractant) binds with the target solute at the feed solution–membrane interface, (*b*) the carrier–solute complex diffuses through the membrane, and (*c*) the solute is released at the membrane-stripping interface. The free carrier then diffuses back to repeat the process (16, 98). Additionally, these studies have elucidated molecular design features that promote solute-selective transport. First, the nonselective diffusion of free solute (i.e., solute not associated with a carrier) should be limited (16). The low solubility of ions in organic solvents helps satisfy this constraint for SLMs. Second, the association–dissociation kinetics between carrier and solute should be rapid such that the reaction may be approximated as being at equilibrium locally. This constraint is satisfied when the reaction rate, $k_{off}^{-1}$, is faster than the rate of diffusion across the thickness of the SLM, $\ell^2/D$. Finally, there is an optimal value of the equilibrium binding constant that enhances solute transport (16, 99). If the interactions are weak and the binding constant is low, the flux due to the selective carriers is negligible. Conversely, if the binding constant is high, the solute binds but does not release readily, so minimal flux enhancements are observed. Significant flux enhancements and high selectivity are realized if the binding constant is tuned adequately (i.e., $K \sim 1/\sqrt{c_f c_r}$) (100). The dependence of the optimal binding constant on the feed and receiving solution concentrations, $c_f$ and $c_r$, respectively, reinforces the need to consider process-scale variables when molecularly engineering membrane materials for solute-selective separations.

Ultimately, despite the robust understanding of transport through SLMs, their limited stability hinders widespread application at the process scale. Namely, the performance of SLMs degrades over time as the carrier molecules leach into the aqueous phase or shear stresses entrain the organic phase in the feed solution. Polymer inclusion membranes (PIMs) attempt to address these issues by embedding the selective carrier (e.g., DEHPA) within a dense polymer film (e.g., cellulose acetate or polyvinyl chloride) that has been blended with plasticizers (e.g., nitrophenyl octyl ether) (101–103). The dense polymer membrane provides durability, and the plasticizer increases the segmental motion of the polymer. However, even with the addition of the plasticizer, carrier diffusion through the polymer is roughly two orders of magnitude slower than diffusion in SLMs. These slow transport dynamics limit the potential of PIMs. Moreover, despite improvements to address carrier loss, this phenomenon remains an issue in PIMs.

Membrane architectures with molecular recognition elements covalently bound to a solid matrix are often designed to circumvent the stability issues associated with SLMs/PIMs. Ideally, if the on–off kinetics related to the solute–ligand interactions are correctly tuned to promote exchange with adjacent carriers, mobility effects may drive solute-selective transport. Structurally, this constraint implies the existence of a percolation threshold and minimum ligand concentration. Above this threshold, the motion of ligands around their tethers allows them to overlap such that solute exchange between ligands could occur. Below this threshold, ligands cannot exchange solute between each other, and diffusion dominates transport. In this membrane architecture, even with overlapping ligands, it is difficult to achieve sufficiently rapid exchange kinetics to realize ligand-based selectivity (16, 99). The difficulty arises because, similar to SLMs, the rate of the association–dissociation reaction should be faster than the diffusion of free solute. However, for tethered ligands, the relevant length scale for diffusion is the distance between ligands (~5–30 Å), resulting in a characteristic rate of diffusion that is much faster than that for SLMs, where the entire membrane thickness (~100 μm) is the relevant length scale. The rapid diffusion rate necessitates a high $k_{off}$ value and appears at odds with the desire to achieve selectivity through large binding constants. Measuring ligand–solute reaction rates within the membrane and identifying an optimal percolation threshold present unique challenges that must be addressed before fixed carrier–facilitated transport can be realized (104).

Emerging studies on dynamic polymer networks aim to overcome the limited mobility of polymer chains that have been implicated as a potential reason for the little success in realizing fixed carrier–facilitated transport. Dynamic polymer networks share properties of thermosets and thermoplastics by using reversible crosslinks (i.e., dynamic bonds) within the polymer backbone (105). The dynamic crosslinks within these materials present new opportunities for engineering the timescales relevant to solute flux. Whereas solute flux in glassy and covalently crosslinked polymers depends on the segmental motion of the polymer, the rate at which crosslinks are broken and formed within dynamic networks provides an additional degree of freedom. Based on computational simulations of a hydrogel, when the size of a noninteracting solute is larger than the mesh size, and crosslinking reactions are fast, the solute cannot diffuse through the network. On the other hand, the network will reconfigure itself around an interacting solute, allowing it to diffuse (106). Realistically, solute–ligand interactions are not instantaneous, and noninteracting solutes may slowly diffuse through the natural reconfiguration of the polymer (107). When the solute size is comparable to the mesh size, reaction rates are particularly important. Here, although increased diffusive fluxes are possible, several experimental systems have presented a decrease in diffusion due to reactive moieties in the polymer matrix (108). Continued experimental efforts on how dynamic bonds influence transport require systematically and independently tuning solute–polymer interactions and system characteristics (e.g., ligand identity or mesh size) (108, 109). In a similar vein, stimuli-responsive materials also offer an additional degree of freedom that may be used to realize solute-selective transport. Specifically, the frequency of a time-varying external stimulus can be tuned to promote solute transport that is faster than diffusion alone (110, 111).

Although membrane architectures with tethered carriers that realize the selective-solute fluxes associated with SLMs have rarely, if ever, been produced, there is precedent for the selective transport of ions that bind to fixed ligands. Polyelectrolyte films possessing iminodiacetic acid groups, capable of achieving a $Cu^{2+}$:$Mg^{2+}$ selectivity of 50, serve as one example (112). Within this system, the selectivity inferred from single-solute experiments is modest. However, the selectivity measured from experiments with multicomponent feed streams is high. Examining the flux of $Cu^{2+}$ and $Mg^{2+}$ best illustrates this idea. In single-solute diffusion experiments, the flux of $Cu^{2+}$ is less than that of $Mg^{2+}$. However, when exposed to multicomponent streams, a partition-driven gating mechanism emerges: The transport of the associating solute (i.e., $Cu^{2+}$) remains the same, whereas the flux of nonbinding solutes decreases (112–114). These results highlight the need for detailed experiments that elucidate the phenomena underlying partition-driven gating, which become apparent only in multicomponent feed streams.

As mentioned above, materials functionalized with strong binding structures will result in the target solute adsorbing to the membrane surface (i.e., they have small $k_{off}$ values). Although this phenomenon typically limits enhancements to permeation rate–based separations, adsorptive separations are well-situated to execute solute-selective separations. Moreover, membranes have several opportunities to advance these processes by altering the sorbent structure and facilitating continuous process operations (115).

Conventional adsorption processes use packed beds filled with porous resins to capture target solutes from a mobile solution that flows through the bed. Mass transfer limitations, associated with diffusion into the beds, result in a portion of the resin remaining unused (115, 116). Adsorptive membranes employ convective flow through the porous material containing the active sites, which reduces mass transport limitations and increases capture efficiency (115). Adsorptive membranes also offer opportunities to alter pore wall chemistry post-synthetically after fixing the sorbent microstructure (117, 118). This independent control over the design of the molecular recognition elements and the sorbent microstructure allows for sorbent design optimization over several length scales (119, 120). For example, lanthanum adsorption was increased by carefully selecting comonomers that enhanced hydrogen bonding between repeat units of polymer brushes grafted onto commercial membranes (121). Similarly, terpyridine-functionalized sorbents were developed to isolate transition metal ions from complex mixtures (e.g., cobalt from lithium, copper from seawater) (119, 122). Ultimately, membranes can be functionalized with molecular recognition elements that draw inspiration from the selective binding pockets of proteins (123–125).

Packed beds and adsorptive membranes require frequent regeneration; therefore, they are operated in batch mode, but continuous processes would be preferred. Continuous countercurrent tangential chromatography uses hollow fiber membranes to facilitate the transition to continuous adsorptive separations by operating binding, washing, elution, and stripping modules in sequence. This membrane-enabled design allows for reduced regeneration frequency, larger residence times, higher purity, and increased productivity compared to conventional packed beds (116). Although membrane materials continue to be developed and improved for solute-specific separations, materials and processes must be designed in concert to maximize separation efficiency.

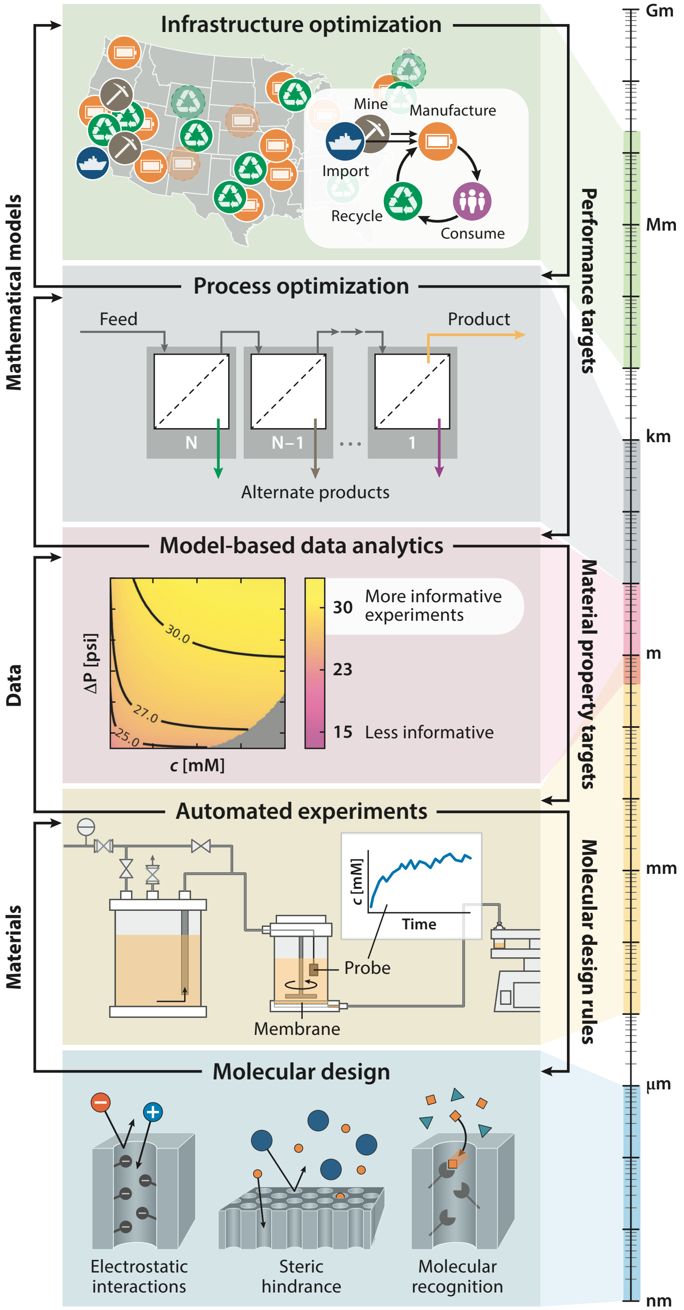
## DIGITAL ENGINEERING ACCELERATES SCALE-UP AND RESEARCH CONVERGENCE ACROSS DISCIPLINES

Realizing the widespread impact of breakthrough membrane technologies for critical mineral processing and recycling requires overcoming diverse barriers across molecular to infrastructure scales. **Figure 5** shows that advances in digital engineering (e.g., material informatics, high-throughput experiments, multiscale modeling, computational optimization, machine learning, and data science) provide exciting new opportunities to accelerate membrane technology development and deployment.

Numerous material platforms with well-defined, tunable nanostructures [e.g., metal organic frameworks (126), covalent organic frameworks (127), and self-assembled block copolymers (128)] allow for the development of membranes with robust frameworks that can be tailored post-synthetically. Tuning the pore wall chemistry offers opportunities to create new materials for targeted separations (65, 117). The many building blocks and pore wall chemistries present a massive phase space of potential materials that must be characterized over a broad range of relevant conditions. As one example, nuanced changes in the pore wall chemistry of copolymer membranes (i.e., changing the spacer arm length between the pore wall and a charged headgroup from an ethylene diamine to a hexamethylene diamine) lead to distinct rejection profiles with increasing $MgCl_2$ feed concentrations (71, 129). As such, identifying materials appropriate for targeted separations and elucidating underlying transport mechanisms will require characterization techniques that can systematically explore a wide range of feed conditions.

Rapidly obtaining informative data will require rethinking the experiments and methods to characterize membrane materials (71, 130–132). For example, a diafiltration apparatus can characterize performance over a range of feed concentrations 10× faster than traditional filtration experiments (71, 130). The concentration space is systematically explored by dosing a diafiltrate solution into the feed solution. Tuning the concentration of the diafiltrate to be greater (or less) than the feed concentration results in an increase (or decrease) in the retentate concentration with respect to time (automated experiments in **Figure 5**) (133). Experiments that enable systematic control over the composition of multicomponent feed solutions offer opportunities to conduct analyses that can identify membrane properties, distinguish between transport mechanisms, and study the interplay between thermodynamics and transport in complex feed solutions.

The gains from experimental automation are multiplied by advances in model-based data analytics. For example, emerging self-driving laboratories combine automation and artificial intelligence to optimize materials rapidly (150–154). However, these laboratories rely on black-box input–output machine learning models that are excellent at optimizing material properties but do not offer physical insights. We argue that model-based design of experiments (MBDoE) is better suited for optimizing separations. Here, the goal is to develop phenomenological mathematical models (e.g., partial differential algebraic equations) to explain transport and thermodynamics in membrane systems. MBDoE accelerates the validation of mathematical models that provide insights into the molecular processes driving separations and facilitate design, optimization, and



*(Caption appears on following page)*

**Figure 5** (*Figure appears on preceding page*)

Resilient supply chains will require molecular to infrastructure optimization of membrane materials and processes. Molecular design: Existing membrane materials separate solutes through steric hindrance and electrostatic interactions, which can be tuned by manipulating the pore size and chemistry, respectively. To separate similarly sized and charged solutes, emerging materials use molecular recognition principles to preferentially transport target solutes through the membrane. Automated experiments: The diafiltration apparatus characterizes membrane materials an order of magnitude faster by exploiting dynamic experiments and automation. Throughout the experiment, a diafiltrate solution is dosed into the stirred cell to explore a predetermined concentration range. The concentration of the retentate and permeate can be monitored continuously using inline sensors. Model-based data analytics: The contour plot shows the information content of a proposed experiment as a function of the diafiltrate concentration and pressure driving force. Model-based design of experiments determines the minimum set of experiments needed to validate lumped parameter or phenomenological models. Process optimization: Mathematical models and optimization identify the most promising opportunities to integrate membrane materials into intensified processes. Infrastructure optimization: Establishing circular economies requires optimizing supply chains and infrastructure, including international trade, domestic mining, recycling, and green manufacturing (e.g., batteries).

scale-up. MBDoE offers a principled way to determine the minimum set of experiments to select, calibrate, and test these models (134–136). First, in model selection mode, MBDoE determines the best experiments to discern between competing models (e.g., two candidate transport mechanisms). Then, in parameter precision mode, MBDoE optimizes the data collection to reduce the uncertainty in physically meaningful kinetic and thermodynamic parameters (e.g., partition and diffusion coefficients) in the best models.

Mathematical models describing membrane performance inform superstructure optimization to design intensified processes with improved energy efficiency and reduced environmental impacts. For example, superstructure optimization recently quantified the trade-offs between purity, selectivity, and the number of stages of optimized diafiltration membrane cascades to fractionate lithium and cobalt (49). This and related studies use process-centric models to help establish quantitative material and device performance targets and provide feedback to shorter time and length scales (133, 137). Similarly, machine learning surrogate models to predict complex transport phenomena (e.g., trained from computational fluid dynamics simulations) or material performance properties can be embedded in superstructure models to facilitate optimization across material, device, and process scales (138, 139). Moreover, modeling can maximize the operational flexibility of intensified processes, thus enabling more rapid responses to feed and market variability (140). Still, there is a research gap in the holistic life cycle analysis of conventional and membrane-based technologies, which can further enhance this process-level analysis.

Infrastructure modeling and optimization enable the integration of new intensified processes into optimized supply chains and circular economies (141). For example, optimization models help inform the siting of battery recycling processes while prioritizing sustainability and circularity (142–145). These developments should be guided by best practices for incorporating life cycle analyses into process and supply chain optimization (146). Similarly, advances in multi-objective optimization can help resolve conflicting opinions across diverse stakeholders while considering the technological, economic, and societal impacts of policy decisions (147).

**Figure 5** is inspired by the success of multiscale modeling and simulation tools to accelerate the scale-up of carbon capture technologies by reducing costs and technical risks to applications (148). **Figure 5** emphasizes the multiscale and interdisciplinary challenges of developing and deploying membrane-based critical mineral separation technologies. These challenges cannot be solved with isolated innovations at only the materials or process scales. Instead, we argue that digital engineering provides frameworks to converge research across diverse disciplines—membrane and materials science, chemical engineering, data and computer science, economics, and policy—to create sustainable circular economies for critical minerals (149).

## CONCLUDING REMARKS

Membrane-based processes could provide economical and environmentally responsible pathways to sustainable critical mineral separations. Relative to current methods, membrane-based processes offer opportunities for process intensification due to their modular nature, limited use of environmentally damaging reagents, adaptability under variable feed conditions, and ability to fractionate input streams into multiple product streams. Existing NF and IEX membranes can separate ions of different sizes and valences, and although improved materials would be helpful to advancing membrane processes, knowledge gaps related to the performance of these membranes in multicomponent feed solutions represent challenging barriers to technology scale-up. Developing physics-based models describing the operation of NF and IEX membranes under these complex conditions can identify optimal applications for current commercially available membranes. This knowledge will also inform the molecular design of new membranes that offer higher selectivity and stable performance in harsh environments. Significant opportunities exist for emerging membrane paradigms that leverage molecular recognition to enable solute-selective platforms capable of separating ions of similar size and valence. Holistic life cycle assessments are necessary to assess the viability of SLMs, which exhibit high selectivities but poor stability, in comparison to membranes with fixed carriers that are more stable but have yet to clearly demonstrate solute-selective transport. Ultimately, the expansive design space available requires convergence across scales and disciplines instead of siloed efforts. In this regard, digital engineering (e.g., self-driving labs, multiscale computational optimization) can accelerate membrane process development and deployment to help realize sustainable economies and secure supplies of critical materials.

## DISCLOSURE STATEMENT

B.W.B. holds a patent and patent application in the space for membranes for water separations. The authors are not aware of any affiliations, memberships, funding, or financial holdings that might be perceived as affecting the objectivity of this review. This presentation was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government, nor any agency thereof, nor any of their employees, nor any of their contractors, subcontractors, or their employees, make any warranty, express or implied, or assume any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government, any agency thereof, or any of their contractors or subcontractors. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government, any agency thereof, or any of their contractors.

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