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Data Science-Enabled Molecular-to-Systems Engineering for Sustainable Water Treatment

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Abstract

Growing social and economic pressures demand technological innovations that enable the widespread usage of unconventional sources of water. These challenges motivate the emerging fit-for-purpose paradigm, wherein water is provided at the precise quality level of the intended application. Unfortunately, to date, fundamental advances in materials and nanotechnology have been slow to advance this paradigm. Using examples from membrane science and engineering, we highlight the critical need to bridge research at the molecular and nano-scales with development at the device and systems-scales to fully realize sustainable fit-for-purpose water technology. Specifically, we present four opportunities for computing and data science to accelerate convergence of sustainable water research: *materials informatics and inverse design*, *model-based design of experiments*, *superstructure optimization*, and *uncertainty quantification*. As such, we highlight opportunities to collaboratively revolutionize molecular-to-systems engineering of sustainable water technologies, but emphasize open communication between data scientists and water-focused researchers using a common vocabulary as a significant hurdle.

Keywords: materials informatics, superstructure optimization, design of experiments, inverse materials design, uncertainty quantification, fit-for-purpose water, membranes

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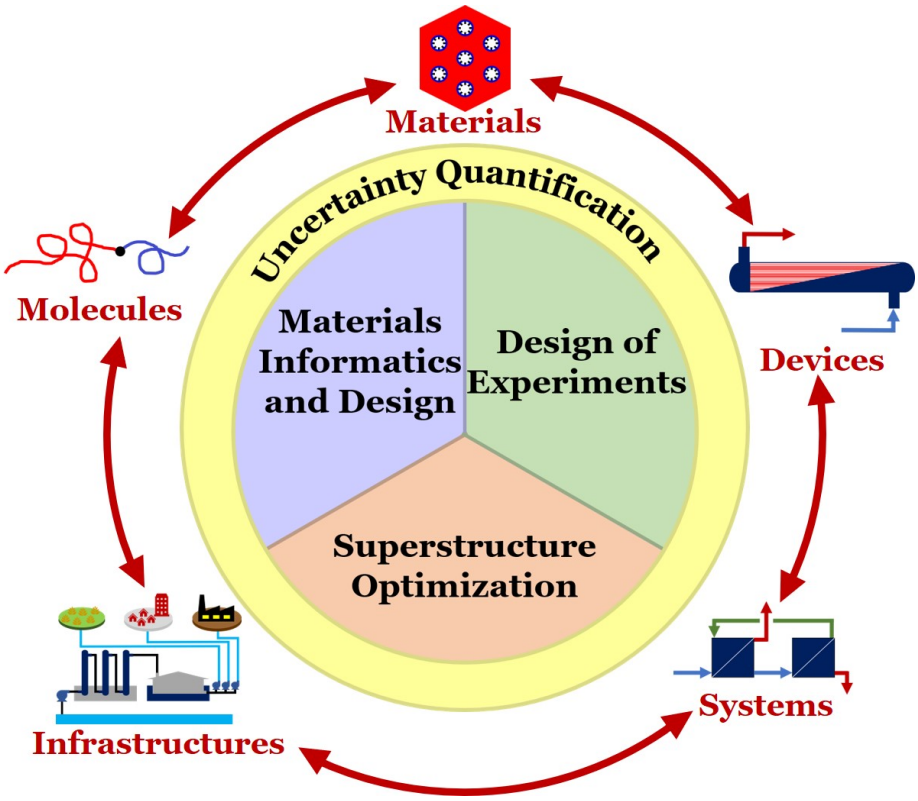

Graphical Abstract

Introduction

Driven by factors such as economic and population growth, aging infrastructure, and increased concerns regarding pollution, water sustainability research is undergoing a paradigm shift to emphasize the highly interconnected and interdependent nature of Earth-water-human systems. Moreover, an increased awareness of the vulnerabilities within existing water supply and management infrastructure has driven interest in the use of non-traditional water resources (e.g., seawater desalination, wastewater reuse) to meet growing demands. As such, several modifications and alternatives to centralized water treatment sys-

tems that produce water of a single, potable quality have been proposed. For example, distributed systems that incorporate regenerative treatment technologies tailored to provide fit-for-purpose water closer to its point of use could be incorporated as part of larger networks [1]. Mihelcic *et al.* [2] broadly define sustainable engineering as "the design of human and industrial systems to ensure that humankinds use of natural resources and cycles do not lead to diminished quality of life due either to losses in future economic opportunities or to adverse impacts on social conditions, human health, and the environment". In this context, sustainable water systems maximize reuse and minimize environmental impact by utilizing a series of treatment processes and recycle loops to produce water at a purity level demanded by the requirements of its users. Moreover, dissolved solutes in waste waters (e.g., nutrients, metal ions) may be viewed as renewable resources that can be recovered. Ultimately, the successful design of these systems will require addressing fundamental questions from the molecular to systems scales related to the development, adoption, and integration of treatment technologies into sustainable networks that ensure robust and resilient infrastructure that can quickly recover from damage or disruption (e.g., natural disasters, equipment failures) [1].

Concurrent with efforts to re-envision the design of the water supply and management infrastructure, advances in chemistry, materials science, and molecular engineering are providing unprecedented abilities to design, characterize, and manipulate materials at the molecular through nanoscales [3]. As such, significant opportunities exist to empower the rational design of materials to positively impact water security by coupling this control to the rigorous design of treatment networks through the development of detailed structure-property relationships. For instance, zirconium metal organic frameworks (MOFs) are excellent candidates for the design of selective sorbents that target the removal of harmful organic contaminants from water treated for direct potable reuse [4, 5]. Moreover, thermoresponsive solvents enable water extraction from high salinity brines using low-cost waste heat as the primary energy input [6, 7]. Due to their ease of operation, modular design, and low energy demands, membrane-

based technologies are exciting prospects for clean water technology [8, 9, 10, 11]. Self-assembled block polymer materials allow for the creation of membranes with pore wall chemistries that are readily tailored to enable solute-specific separations and detection. For example, membranes with pore walls tailored to detect and capture metal ions, if appropriately designed, could be used for resource recovery or remediation efforts [12, 13, 14]. Nanocomposite membranes, which are fabricated by incorporating nanomaterials into the matrix of conventional membrane structures, can be designed to promote the inactivation of microorganisms that lead to disease and biofouling [15]. Alternatively, nanocomposite membranes can be designed to enable localized solar-thermal heating that enhances membrane distillation processes [16]. While the promise of these materials is exciting, transformative advances for sustainable water have been slow to manifest in practice. Due to their time- and resource-intensive natures, the empirically-driven, heuristic methods that guide most efforts to enhance material properties and device performance are one impediment to realizing this potential. Typical workflow are often narrowly focused on a single, fixed system design and do not incorporate feedback from rigorous systems analyses and process synthesis optimization.

Accelerating materials discovery necessitates a move away from classical Edisonian methods to principled and data-driven frameworks that can guide material design and process synthesis to overcome the gaps in knowledge that inhibit the translation of new materials and devices from the laboratory scale to sustainable water treatment technologies. While there is consensus that materials enabled solutions will play an important role, realizing the goal of developing sustainable water technologies and resilient water management infrastructure requires inputs from several fields of study. This critical review offers the perspective that data-science enabled paradigms can precipitate the development of materials and technologies for sustainable water by accelerating research at intersection of materials science, computer science, civil and environmental engineering, mechanical engineering, and chemical engineering. In Figure 1, we elaborate the multiple length scales of molecules-to-systems design along with

four data-driven tools, namely, materials informatics and inverse design, design of experiments, superstructure optimization and uncertainty quantification, that can be applied at these various scales.

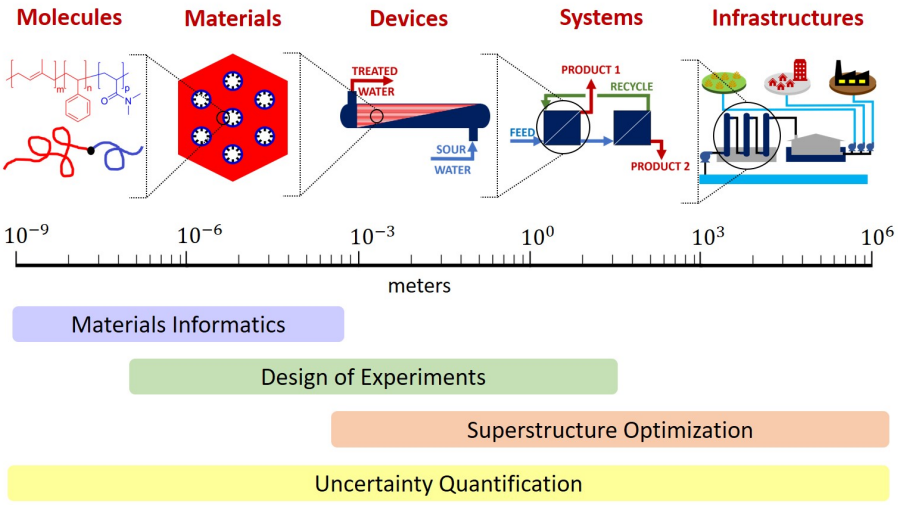

Figure 1: In the bottom-up approach to sustainable water engineering, new (macro)molecular structures enable novel materials with desired properties that are incorporated into devices for target applications. These devices are then integrated into process networks wherein the system topology enables non-traditional applications such as nutrient recovery and wastewater treatment which form an essential part of the water distribution infrastructure in the fit-for-use paradigm. Computational and data science tools can accelerate this process to facilitate top-down analysis and discovery of materials. In the top-down approach, *superstructure optimization* is used to design resilient infrastructures and novel system configuration for specific water purification and solute recovery applications. These designs dictate targets for material properties that guide the inverse design of materials. Likewise, bottom-up *materials informatics* facilitates rapid screening in the context of fully optimized systems and infrastructure. *Design of experiments* helps systematically test scientific hypothesizes and accelerate technology scale-up. Multiscale *uncertainty quantification* and propagation unite all the computational and data science tools into a holistic molecules-to-infrastructures framework.

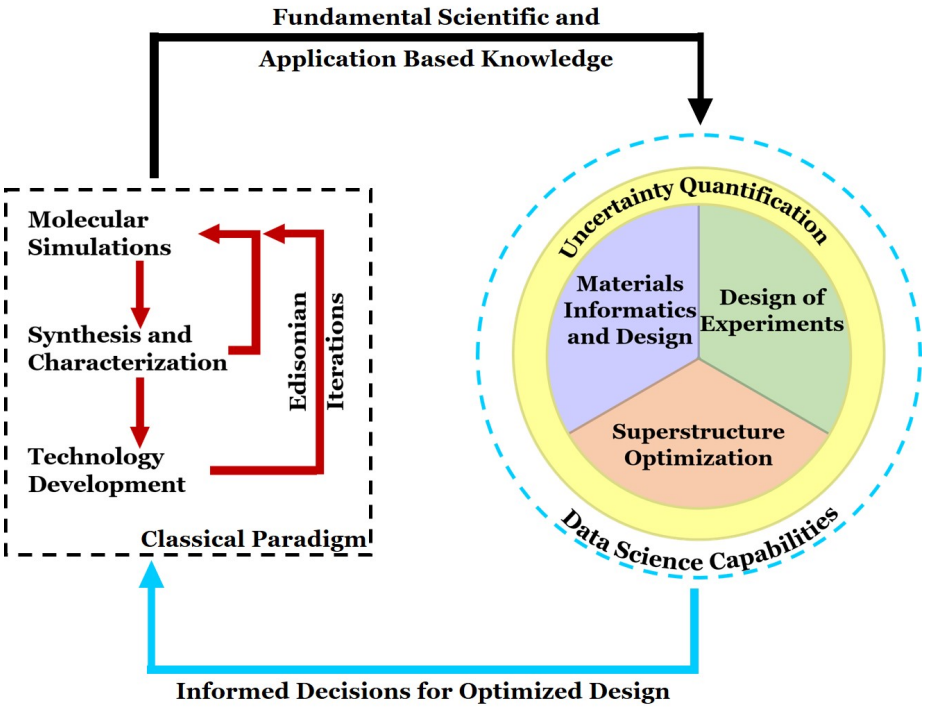

Figure 2: The proposed paradigm integrates the classical workflow with four data-science enabled capabilities to optimize the materials discovery process by maximizing the amount of useful information gained from each iteration and enabling simultaneous multiscale analysis.

Opportunity 1: Materials informatics and inverse design

We foresee the convergence of materials informatics, Bayesian optimization, and inverse design enabling two to three orders of magnitude acceleration in identification of promising membrane materials tailored for sustainable water technology. The goal of inverse design is to computationally predict (macro)molecular and/or self-assembled structures that achieve target material properties. Although empirical structure-property relationships exist for some polymer structures [14], the fundamental understanding of molecular interactions and transport mechanisms that govern solute-specific separations are still rudimentary [17]. Materials informatics leverages massive online datasets to automatically learn structure-property relationships that are vital to solve the

inverse problem of materials discovery [18]. We anticipate the imminent confluence of materials informatics with physical and synthetic chemistry will lead to new insights and more accurate structure-property relationships enabling automated frameworks that expedite the engineering of polymer nanomaterials. In Figure 2, we show the sequential and heuristic nature of the conventional materials discovery workflow and contrast it with the integrated nature of data-science tools highlighting the benefits that each paradigm can gain from the other.

The nearly limitless design space of candidate materials cannot be enumerated with conventional high throughput computational screening methods [19, 20]. Instead, in materials informatics, a surrogate or machine learning model is constructed to predict material properties (e.g., solubility) from input design variables (e.g., molecular structure). Adaptive design techniques refine the surrogate model through continuous (re)learning and postulate new materials to synthesize or simulate using the surrogate model. A popular technique is to use Bayesian optimization methods, in which domain specific knowledge (e.g., existing structure property relationships) and observed data are incorporated into the prior distribution of the surrogate model [21]. The computationally inexpensive surrogate model enables optimization of experimental conditions that balance *exploration* (sampling regions of high uncertainty) and *exploitation* (sampling regions with best predicted material performance) when selecting future experiments. New observations are leveraged to improve the surrogate model using Bayes rule, making the procedure adaptive (self-learning). Adaptive designs have been shown to outperform pure exploitation approaches in several studies [22] with successful applications including the study of self-assembled nanoparticles [23, 24], therapeutic drugs [25], and high strength alloys [22].

Despite their potential to accelerate technology development, data-driven inverse design techniques pose several challenges and opportunities. Thus far, many successful implementations of adaptive designs are with either limited design spaces (e.g., ternary alloys) or crystalline materials with well-defined, equilibrium structures. In contrast, structure-property relationships for amor-

phous soft materials such as polymeric and nanocomposite membranes are not as readily available or as easily defined. Polymer membranes, in particular, have extremely large design spaces because they are processed far from equilibrium and therefore, possess non-equilibrium structures. New mathematical descriptions grounded in physical and chemical understanding are needed to encode all of the design choices for soft, polymeric materials. In some cases, detailed molecular simulation may be needed to elucidate the self assembly process and ultimately predict structure and properties of soft materials [26]. This trade-off between computational expense and molecular-scale detail invites novel mutlifidelity adaptive design methods to guide inverse materials design. Finally, the water treatment technologies in which these materials are deployed are often multifaceted and utilize a combination of phenomena to achieve their goal. This broad design space necessities advances in multiobjective optimization to navigate competing design goals (material targets) and assimilate heterogenous data from many sources.

Opportunity 2: Model based design of experiments accelerates hypothesis driven discovery

A fundamental challenge in scientific discovery is designing experimental campaigns that maximize useful information gained to examine governing hypotheses [27]. We foresee model based design of experiments (MBDOE) facilitating greater insight into the thermodynamic and transport phenomena governing membrane-based processes. MBDOE is a statistical technique to achieve a user-defined goal such as discriminating between rival models or improving estimated parameter accuracy with the fewest number of experiments [28]. Classical design of experiments techniques (e.g., multi-level factorial, partial factorial designs) help establish empirical models (e.g., polynomial response surfaces), which although predictive, rarely offer insights into fundamental scientific phenomena. In contrast, MBDOE frameworks directly consider differential and/or algebraic equations grounded in scientific and engineering fundamentals (e.g.,

conservation laws, thermodynamics) in their formulation. MBDOE techniques are popular in biology, pharmacology, and reaction engineering, as these experiments are often time- and resource-intensive. For example, Laínez-Aguirre *et al.* use a fully Bayesian MBDOE framework to estimate nonlinear differential algebraic pharmacokinetics models which enable dosing regimens optimized to individual patients [29]. In another recent example, Han *et al.* [30] use MBDOE to distinguish between competing kinetics models for chemical-looping combustion. Despite the fact that similar challenges exist in sustainable water treatment (e.g., discerning fouling mechanisms, identifying degradation pathways of contaminants), MBDOE techniques have seldom been leveraged in this arena.

As highlighted in Opportunity 1, the transport, thermodynamics, and reaction mechanisms that enable water treatment technologies are often not sufficiently understood. We believe it is possible to postulate model collections where each model corresponds to specific scientific hypotheses about the dominant physical and chemical phenomena. MBDOE then facilitates design of multifaceted experimental campaigns to discern the most probable subset of models (hypotheses). This proposed paradigm is especially powerful in the elucidation of regimes where a single mechanism dominates and the identification of conditions where transitions between mechanisms occur. For example, it could potentially be utilized to elucidate where transport through membranes transitions from being governed by a solution-diffusion mechanism to a pore flow dominated regime. Moreover, MBDOE can enable the design and analysis of high throughput transient experiments, thereby eliminating the time required for a test system to reach equilibrium.[31] This capability may be particularly useful when testing new technologies against realistic feed solutions. Often materials characterization is executed using idealized solutions that contain a single dissolved component. In practice, however, water treatment technologies are challenged by complex, multi-component solutions whose composition varies with time. As such, there is a critical need to assess material and device performance in a broader spectrum of feed solution conditions. In this regard, the

proposed capabilities of MBDOE would accelerate development of sustainable water technologies that can robustly operate over wide ranges of conditions.

Opportunity 3: Superstructure optimization enables rapid bottom-up and top-down analysis

We envision that superstructure optimization can provide a mathematical framework to leverage structure-property relationships (Opportunity 1) and validated transport and thermodynamic models (Opportunity 2) for holistic molecular-to-systems design that encompass all aspects of sustainable water. Superstructure optimization is a classical paradigm in process systems engineering with ubiquitous applications including water distribution networks [32, 33], water supply chains [34], and sensor placement [35]. First, the modeler postulates a superstructure at the desired length scale that encodes all possible system configurations. Next, an optimization problem is formulated over the superstructure. For example, minimize water production cost by searching over all feasible combinations of design choices (e.g., selection and size of equipment, flow rates). Finally, the optimization problem is solved numerically, resulting in one (or more) designs with optimal topology (discrete decisions) and operating conditions (continuous decisions) identified. Often, superstructure optimization elucidates novel system topologies. For example, Du *et al.* [36] discovered permeate split designs for a single-feed, multi-product seawater reverse osmosis desalination network using superstructure optimization, thus demonstrating how commercially-available materials, if optimally integrated into devices and systems, can overcome operational challenges such as boron removal. Yenkie *et al.* [37] use superstructure optimization to calculate quantitative performance thresholds that define when various separation pathways minimize production costs.

Extending established top-down superstructure optimization methods to encompass emerging fit-for-purpose paradigms and the additional constraints needed to address resource recovery and water reuse can help to focus research

efforts into these highly-integrated, complex systems on the most impactful areas[38]. In this domain, there is a great need to understand the basic cost and performance drivers [39] for new sustainable water technologies in the context of existing or future infrastructure, regulations, and public opinion with many stakeholders [40]. Superstructure optimization is often overlooked during materials development. However, as shown in Figure 3a, emerging efforts in this arena are focused on establishing top-down guidance at the device level by identifying material property targets needed to enable novel process configurations [36, 41, 42]. Moreover, top-down approaches are beginning to span the divide between the materials and systems scale by embedding empirical correlations that guide materials selection within the optimization framework. For example, the Robeson plot, which quantifies the trade-off between permeability and selectivity for membranes was considered when optimizing the design of binary gas separation systems [43, 44, 45]. Incorporating these structure-property relationships into the optimization framework can both quantify the relative importance of competing material properties, as seen in Figure 3b, and elucidate the potential for performance gains at the systems level from technological breakthroughs, i.e., shifting the Robeson plot. As such, superstructure optimization frameworks of the future could inform both *how to design* nanostructured materials and *how to integrate* them into resilient infrastructures with many competing objectives.

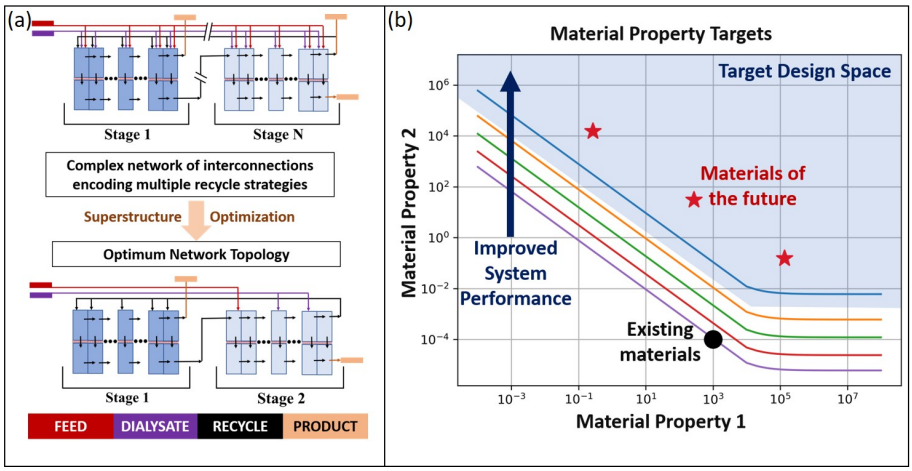

Figure 3: **(a) Superstructure Optimization:** In superstructure optimization, a comprehensive superstructure encoding all possible network configurations is decomposed into a network with optimum topology. For example, a full superstructure encoding multiple recycle strategies for a membrane separation cascade is shown at the top. A solution showing optimum strategies for feed injection, product withdrawal and recycle is obtained by eliminating streams whose flow rates are zero and is shown in the bottom of the figure. The optimum configuration is found by performing a sensitivity analysis to identify membrane properties needed to outperform existing technology. **(b) Material Property Targets:** Targets for competing material properties are defined by operational (process) and consumer needs. Materials of the future are tailored for target applications such as heavy metal removal, and can be designed by significantly improving any one of the two competing material properties (vertical or horizontal movement into the target design space), or by modest improvements in both material properties (diagonal movement into target design space) which leads to the overall improvement of system performance.

Superstructure optimization is also well-positioned to guide molecular engineering via bottom-up analysis or inverse design. For already characterized materials, rigorous superstructure optimization enables rapid comparison against competing technologies in the context of fully optimized systems and can identify potential applications for materials. Recent efforts propose superstructure optimization formulations to design crystalline material structures, such as MOFs, based on the constraints imposed by process separations [46, 47]. Extending this bottom-up analysis to soft materials, such as polymeric membranes, remains a challenge since phenomena governing solute-specific separations is not well understood. Nevertheless, we anticipate materials informatics (Opportunity 1) and MBDOE (Opportunity 2) will lead to new insights and more accurate structure-property relationships needed for direct molecules-to-infrastructure optimization. Furthermore, we foresee opportunities to combine molecular simulations and superstructure optimization to enable high-throughout screening of emerging materials using systems-scale metrics (e.g., separation selectivity, energy usage, product cost) as benchmarks for performance.

Opportunity 4: Multiscale uncertainty quantification derisks technology development

Fully realizing the promise of molecular-to-systems engineering to design efficient, reliable, sustainable, and resilient water infrastructures necessitates managing uncertainty across vast length and timescales. Optimization under uncertainty is a well-established paradigm to design water networks that are resilient to supply-demand variations, equipment failures, rare events, and other sources of infrastructure-level uncertainty.[48] Although recent work emphasizes data-driven statistical techniques to define uncertainty sets (e.g., probability distributions) [49, 50], data for water infrastructures are often difficult to obtain or incomplete and erroneous. At the materials scale, all encompassing databases of properties along with quantified reliabilities are difficult to develop due to the resource intensive nature of data accumulation and validation [19]. Experimen-

tal inaccuracies and incomplete models further obscure data collected. These challenges induces uncertainty in the device and systems models that directly inform infrastructure design. We see a great need and opportunity to extend optimization of resilient water networks to address these challenges and encompass all aspects of sustainable water. Recently, Bhat *et al.* demonstrated how Bayesian uncertainty upscaling, which encompasses modeling fundamental thermodynamic and transport phenomena, bench-scale demonstrations, and superstructure optimization under uncertainty, can derisk CO_2 capture technologies. [51] Similarly, establishing molecules-to-infrastructure uncertainty quantification (UQ) frameworks can realize new synergies between Opportunities 1 - 3. For bottom-up analysis, UQ offers the ability elucidate how a new material can impact the performance of water networks while directly considering uncertainty at all technology development levels. From a materials screening perspective, UQ can determine when a new materials is conclusively superior or inferior to alternatives. Moreover, UQ can illuminate where to focus scientific and engineering efforts to reduce uncertainty to improve overall systems performance.

We envision suistainable water applications driving new methodological advances in multiscale UQ. In Figure 4, we sketch an integrated UQ workflow and specifically highlight connections with Opportunities 1 - 3. For example, iterations between molecular simulations and experiments are at the core of the existing materials discovery paradigm (Figure 2). Emerging UQ frameworks enable rigorous calibration of force field parameters and assessment of predictions from molecular simulations [52]. We see opportunities to develop new multi-fidelity design of experiments algorithms to co-optimize allocation of computational and physical resources in order to establish systematic feedback loops between molecular simulations and physical experiments. Supported by rigorous statistical analysis, molecular simulations and physical experiments provide rich heterogeneous datasets with uncertainty estimates to derive (differential) algebraic surrogate (i.e., reduced order, timescale bridging) models that enable computationally tractable optimization under uncertainty [53, 54]. Here, there are significant opportunities to extend the surrogate modeling paradigm

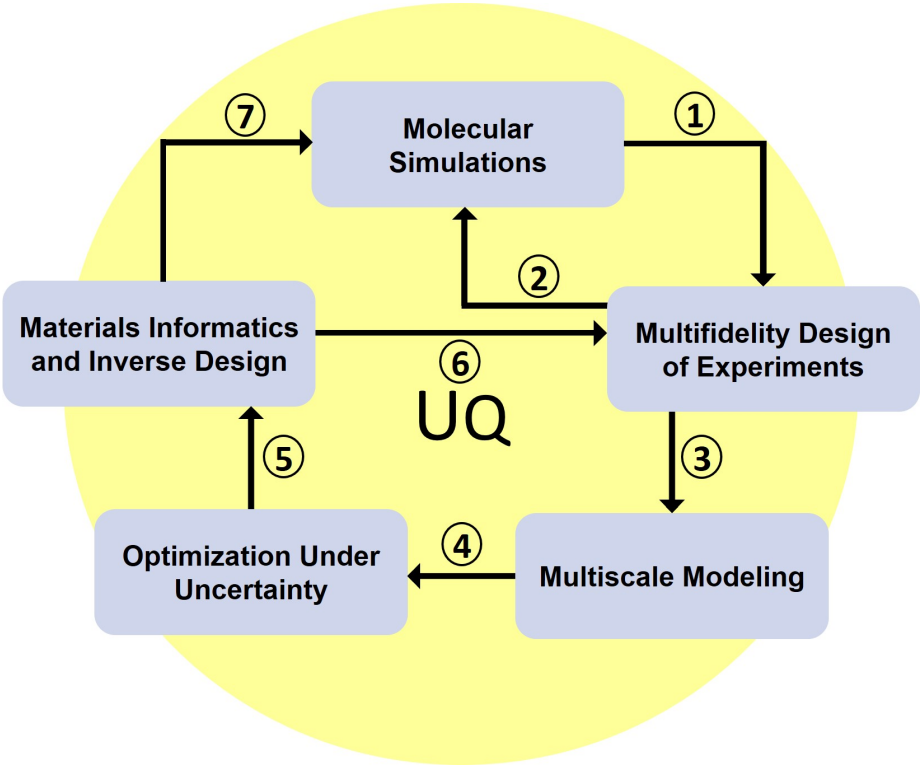

Figure 4: Uncertainty quantification (UQ) unifies all aspects of sustainable water engineering into comprehensive molecular-to-infrastructure design framework. Together, molecular simulations and UQ enables accurate force-field selection and calibration. This provides molecular structure and material property predictions with quantified uncertainties ① to inform laboratory experiments and DOE. Likewise, multifaceted laboratory experiments provide data ② for molecular simulation validation, creating a feedback loop. Multifideltiy design of both laboratory and computational experiments provides rich heterogeneous data with uncertainty estimations ③ for multiscale model reduction. This then provides tractable surrogate (low fidelity) models with quantified uncertainty descriptions ④, which are inputs for superstructure optimization under uncertainty. These optimization problems elucidate possible applications for new materials, material property targets, and device, system and infrastructure designs that are robust to uncertainties (variable feed compositions, rare event, etc.). These results, especially property targets, ⑤ are inputs for inverse design of materials. Adaptive design of experiments (e.g., Bayesian optimization) then proposes tailored materials, ⑥ and ⑦, for synthesis, laboratory characterization, and computational exploration.

to hybrid models, which combine physics-informed equations with a data-driven component that quantifies model form uncertainty [51, 55]. Using these uncertainties, superstructure optimization can provide quantitative material property and cost targets that serve as input for inverse materials design. Critically, this vision necessitates integrating innovations in data science and UQ with fundamental scientific and engineering principles to accelerate sustainable water technologies faster than both fields working separately.

Conclusions

In this review, we highlight four opportunities for emerging data-science frameworks to establish new paradigms for sustainable water technologies: materials informatics and inverse design, model based design of experiments, superstructure optimization, and uncertainty quantification. Many engineering challenges in the sustainable water domain are likely intractable for existing computational paradigms, but provide timely and impactful applications to motivate methodological advances in data and computer science. Data-driven tools cannot alone revolutionize the field. Instead, we foresee collaborative efforts that combine domain specific knowledge with data science paradigms to realize holistic molecules-to-infrastructures engineering frameworks that usher in the next era of materials for sustainable water enterprises and beyond.

Acknowledgements

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Conflict of interest statement

Nothing to declare.

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