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Perspective

Toward smart carbon capture with machine learning

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SUMMARY

Machine learning (ML) is emerging as a powerful approach that has recently shown potential to affect various frontiers of carbon capture, a key interim technology to assist in the mitigation of climate change. In this perspective, we reveal how ML implementations have improved this process in many aspects, for both absorption- and adsorption-based approaches, ranging from the molecular to process level. We discuss the role of ML in predicting the thermodynamic properties of absorbents and in improving the absorption process. For adsorption processes, we discuss the promises of ML techniques for exploring many options to find the most cost-effective process scheme, which involves choosing a solid adsorbent and designing a process configuration. We also highlight the advantages of ML and the associated risks, elaborate on the importance of the features needed to train ML models, and identify promising future opportunities for ML in carbon capture processes.

INTRODUCTION

Climate change, resulting from the emission of greenhouse gases, especially carbon dioxide (CO_2), has become a major concern in recent years.[1–3] Carbon capture technologies can be considered an effective strategy for reducing CO_2 emissions. Carbon capture is broadly recognized as a range of processes for removing CO_2 at the point source (e.g., industrial flue gas) or from the atmosphere through direct air capture.[4–6] Once CO_2 is captured, it can be either permanently stored (e.g., underground) or used to produce high-value products, such as fuels or specialty chemicals in a process known as carbon utilization.[7,8] Carbon capture processes are being developed and are an immediate priority for achieving emission mitigation goals.

State-of-the-art technologies for CO_2 capture involve an absorption route, in which an absorbent agent captures CO_2 from a mixed gas stream, followed by a thermal-stripping process, in which pure CO_2 is released and the absorbent agent is regenerated.[4,9] Most thermal-based capture systems use an amine, such as monoethanolamine (MEA), diethanolamine (DEA), or methyldiethanolamine (MDEA), although other absorbents, including potassium carbonate (K_2CO_3) and amino acids (such as glycine salts),[10–12] have also been widely investigated. In recent years, ionic liquids (ILs) have attracted significant attention as alternative absorbents that potentially offer a lower vapor pressure and less thermal degradation compared with amines.[13–15]

In addition to absorption-based carbon capture, adsorbent-based processes have also been widely investigated because they promise to be more energy efficient and versatile for a variety of carbon capture applications. During an adsorption-based separation, CO_2 is selectively adsorbed on the surface (or within the matrix)

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of an adsorbent substrate, which can subsequently be regenerated, e.g., by a thermal or pressure swing.[16] Adsorbent-based CO_2 capture processes have been considered for both pre- and post-combustion conditions as well as in direct air capture settings.[17,18] Conventional adsorbents include activated carbon, zeolite, silica gel, and metal oxides.[19,20] In recent years, metal-organic frameworks (MOFs) have been studied widely as a promising alternative adsorbent for CO_2 capture, with advantages of high selectivity, high capacity, large specific surface area, and structure-property tunability; thus, there is a broad design space for synthesizing a wide range of these materials.[21–24]

Advances in machine learning (ML) are having a large effect in many fields of study, including chemistry, physics, and materials science,[25–28] unleashing the substantial potential of ML as a powerful emerging tool for carbon capture applications as well. ML methods have enabled researchers to design, test, and improve various aspects of the process that are computationally (using theoretical calculations) or experimentally time consuming and expensive. The use of ML for carbon capture processes is still emerging, and most investigations have been conducted only in the past few years.

The primary goal in implementation of ML for carbon capture is to design and execute process schemes to effectively separate CO_2 from a gas mixture (e.g., $CO_2/N_2/O_2$) with a minimum energy penalty and cost. ML methods have been implemented successfully in both absorption- and adsorption-based processes, at the molecular and the process level, to overcome challenges that these approaches are currently facing. In this perspective, we discuss how ML has been adapted to predict the thermodynamic properties of CO_2-absorbent chemistry, such as the solubility, to facilitate the discovery of alternative absorbents. In addition, we elaborate on how ML methods have been employed to optimize the absorption-desorption scheme, aiming to minimize the overall energy penalty of the process and to maximize the CO_2-capture rate, which results in reducing the capture cost. For adsorption, ML techniques have been implemented to explore many options to find the most cost-effective process scheme, which involves choosing a solid adsorbent and designing a process configuration. We highlight the need for the development of efficient methods offered by ML for rapid screening and for exploring those options. We also identify promising future opportunities for ML in carbon capture processes, including the corresponding risks and considerations in its implementation, for both absorption- and adsorption-based operations. Overall, we provide an overview guide for ML in carbon capture, which should be helpful for scientists working on either the computational or the experimental frontiers of this topic and who may be interested in implementing this powerful tool. Figure 1 presents a schematic of the ML implementation at various scales for a carbon capture process based on different sources. In addition, the logic behind the construction of an ML model as a data-driven tool, the model-training step, and its subsequent applications for prediction and/or optimization purposes are illustrated. Detailed information regarding the logic governing the construction and evaluation of ML tools can be found in the literature.[29–31] Specifically, comprehensive reviews are available on how to build machine-learning models[31] and how to use them to design porous materials.[32]

MACHINE LEARNING FOR CARBON CAPTURE

ML tools have been employed to assist in the development of carbon capture processes on several fronts. In this section, we discuss how ML methods have been implemented in absorption- and adsorption-based processes, both at the molecular

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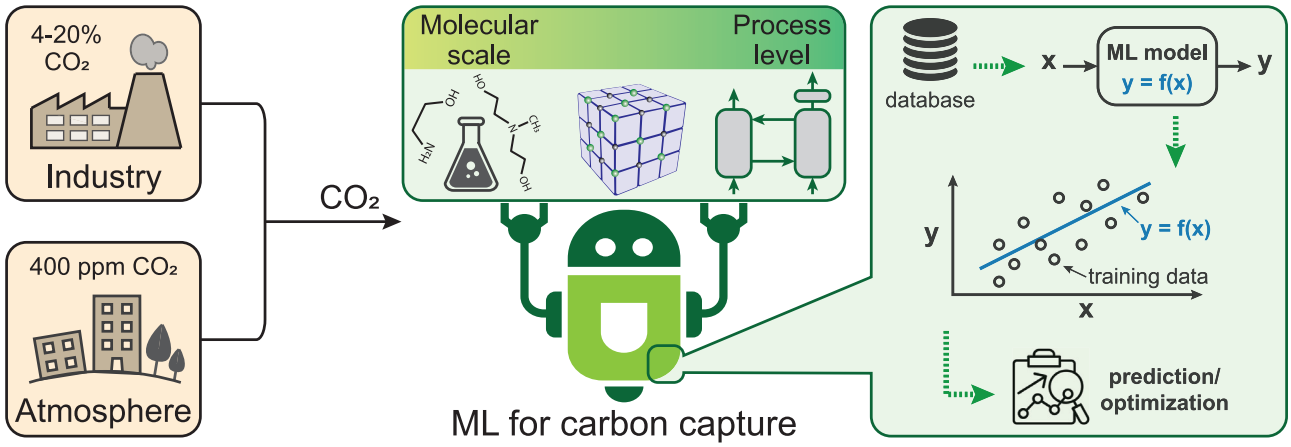

Figure 1. Schematic of ML implementation at various scales for carbon capture processes

CO_2 can be captured from a point source (e.g., industrial flue gas with 4%–20% CO_2) or the atmosphere (~400 ppm CO_2). ML can be applied to improve the capture process at either molecular- or process-level scales. In a typical ML workflow, data collected either experimentally or through simulation are used to train an ML model, which can later be used for prediction and/or optimization purposes.

and the process levels. These implementations aim to overcome challenges currently facing those processes to facilitate further developments to achieve effective, large-scale carbon capture.

Absorption-based carbon capture

Predicting the thermodynamic properties

In the development of absorbent-based carbon capture processes, the thermodynamic properties of the CO_2-absorbent system have a decisive role. Among those properties, the CO_2-equilibrium solubility is of particular interest and is used to evaluate the performance of absorbents in carbon capture processes. Several thermodynamic models have been established to evaluate CO_2 solubility in an absorbent under various operating conditions, ranging from classical thermodynamic methods, based on the vapor-liquid equilibrium (VLE) theory (such as the nonrandom two-liquid [NRTL] model) to molecular dynamics.[33,34] However, limitations remain regarding the accuracy and range of these prediction models, hindering their subsequent applicability for predicting CO_2 solubility in diverse absorbents. As a promising alternative to thermodynamic approaches, ML models have been employed to predict the equilibrium solubility of CO_2 in a wide range of absorbents. These algorithms can effectively reflect the complexity of a system and the inherent relationship between the features and the solubility with high confidence and precision. These features include operational temperature and pressure, CO_2 partial pressure, and absorbent-related properties, as be discussed below.

As the most frequently used absorbent in large-scale carbon capture processes, amines have received the most attention. ML models have been developed to predict the CO_2 solubility in a wide range of CO_2–H_2O–amine(s) systems. All types of amines, either individually or in a mixture, including primary and secondary amines[35–37] (which offer rapid absorption kinetics) and tertiary amines[38–40] (which offer high absorption capacity), have been investigated. ML models have been constructed and trained based on experimental data for the solubility of CO_2 in amines, as available in the literature. The common features employed in these models include temperature, CO_2 partial pressure, and amine concentrations.[35,41,42] Although the models developed thus far have been quite accurate, the applicability of the models remains questionable. The key features missing from the developed

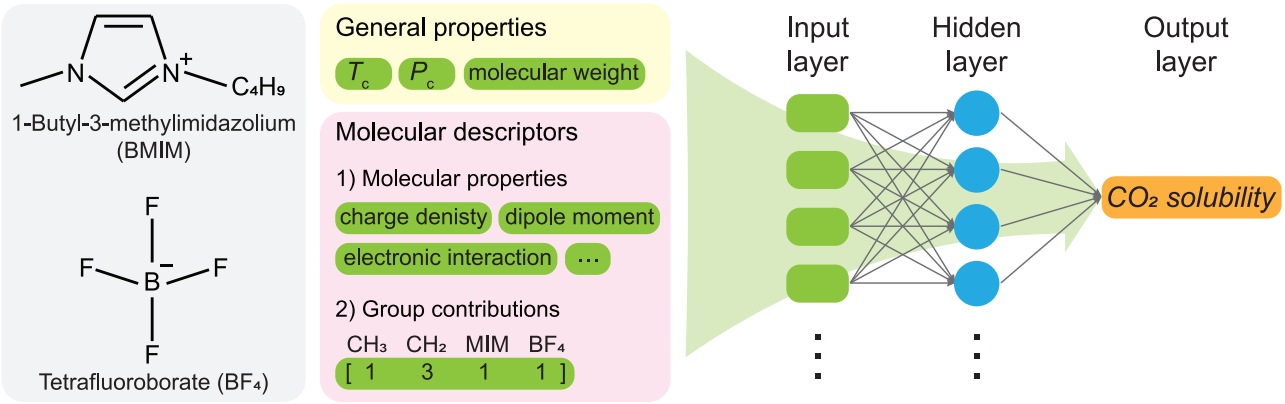

Figure 2. A demonstration of ML implementation to predict CO_2 solubility in an IL

The CO_2 solubility of an IL can be predicted through an appropriate ML method, e.g., using an artificial neural network (ANN). This illustration indicates various types of input features used to develop a model for 1-butyl-3-methylimidazolium tetrafluoroborate (BMIM BF_4) as an IL case example. General properties (including the IL critical temperature [T_c], pressure [P_c], and molecular weight) and molecular descriptors can be used as the model input. Two types of molecular descriptors have been investigated, including molecular properties, such as charge density, and group contributions (occurrence of functional groups).

models are those related to the amine itself, such as the amine type and molecular structure. For example, tertiary amines generally have higher capacities compared with primary or secondary amines at the same concentration; however, if the model does not consider the amine type, it cannot accurately predict the solubility. One of the key features of these models is the ability to reveal the relative importance of each parameter. With an amine as the absorbent, the CO_2 partial pressure is often reported to have the most-significant effect on equilibrium solubility.[37,41,42] In future developments, in which the type of amine and its molecular structure are incorporated into the model as input features, the relative importance of the parameters must be re-evaluated.

ILs have drawn significant attention as a promising alternative agent to amines for carbon capture, because they feature low vapor pressure, high thermal stability, and structure tunability.[43,44] An IL is a salt consisting of a cation (e.g., 1-butyl-3-methylimidazolium [BMIM]) and an anion (e.g., tetrafluoroborate [BF_4]), whose melting point is relatively low (e.g., room temperature), resulting in a liquid salt. Because of the many cations and anions, ILs are highly tunable; however, it is challenging to rationally select and design the most suitable candidate for carbon capture. To address that issue, ML methods can be implemented to predict the key thermodynamic properties of ILs, such as the CO_2 solubility, which is essential for narrowing the list for an effective IL-based carbon capture system.

Various groups of features are considered as the model input to predict CO_2 solubility. A series of investigations considered the general properties of ILs, including their critical pressure and temperature, their molecular weights, and their key process parameters, such as the operational temperature and pressure[45–47] (Figure 2). Although the developed models can predict the solubility values in a given IL, they lack deep physical insights regarding the chosen features. The CO_2 solubility within an IL is affected by various parameters, especially those related to the chemical structure of the IL. The chemical structure of an IL directly affects key properties, such as intermolecular interactions and charge density, which must be considered when predicting CO_2 solubility. Therefore, it is necessary to include the chemical structure of an IL as an input feature, rather than relying solely on general properties,

to develop a model that is applicable for predicting the CO_2 solubility in a wide range of ILs.

To address that issue, several models have been developed to predict the CO_2 solubility by considering the IL chemical structure as an input feature. In this context, molecular structures are first broken down into a series of numerical values using analytical methods, thus describing its relevant chemical and physical properties.[48] These numerical values, which are known as molecular descriptors, are subsequently used as input features to develop an ML-based model, ensuring that the IL molecular structure is effectively considered when predicting the CO_2 solubility.

In general, two types of molecular descriptors are used: those related to the molecular properties, and those related to group contributions. As an example of the former, Venkatraman and Alsberg[49] developed an ML model to predict CO_2 solubility, in which 100 molecular properties (such as charge density, dipole moment, electronic interaction, among others) were used as descriptors for each IL to construct the model (Figure 2). In addition to creating a precise model to predict solubility values, they found that descriptors related to the geometrical and physicochemical properties of both cations and anions, particularly those relevant to intermolecular interactions, have the greatest effect on the CO_2 solubility in a given IL. For group contributions, the molecular descriptors represent the occurrences of functional groups (e.g., OH) in the molecule. This approach potentially offers a simple route because the input feature matrix is relatively easy to construct; the value assigned to each functional group is the occurrence frequency of that particular functional group. For example, for BMIM BF_4, the input matrix is [CH_3: 1, CH_2: 3, MIM: 1, BF_4: 1] (Figure 2). Based on that strategy and a comprehensive dataset containing more than 10,000 CO_2-solubility data points for ILs at different temperatures and pressures, an ML-based model was developed that precisely predicts solubility values.[50] These findings highlight the importance of including the IL chemical structure when developing an ML model of CO_2 solubility.

In addition to amines and ILs, the equilibrium solubility of CO_2 in other absorbents has been predicted using ML algorithms. In this context, the solubility values in solutions of various amino acids, such as sodium glycinate[51] and potassium lysinate,[52,53] and in inorganic solvents, such as trisodium phosphate solutions,[54] have been modeled using various ML algorithms. The CO_2 partial pressure, temperature, and absorbent concentration have been considered as input features to construct the model; however, as mentioned earlier, this approach may result in a lack of deep insights because these models do not consider the chemical nature (and related parameters) of the absorbent. CO_2 solubility in physical solvents (as opposed to water-based chemical absorbents) has also been predicted by a descriptor-based ML model. Organic solvents, such as methanol, acetone, and ethylene glycol, were considered, wherein the input descriptors included the organic structural and bond information, thermodynamic properties, and experimental conditions.[55] Using those descriptors, intrinsic relationships between the general properties of physical solvents and their associated CO_2 solubility were well fitted with an appropriate ML algorithm.

In addition to the solubility, other important thermodynamic properties of the CO_2-absorbent system that affect the overall performance of the carbon capture process have been predicted using ML methods. Among these properties, the density, viscosity, toxicity, and surface tension of the CO_2-absorbent system have been well correlated to operational parameters, such as the temperature and CO_2 partial

pressure, as well as to the absorbent properties (e.g., using group contributions).[52,56–58] These essential thermodynamic properties should be considered carefully for large-scale operations. For example, a high viscosity of a CO_2-absorbent mixture leads to an increase in the energy penalty associated with the solution circulation between the absorption and desorption columns. Therefore, ML methods that can accurately predict the viscosity for a wide range of CO_2-absorbent systems would be highly beneficial for selecting the most suitable absorbent and operational conditions. Foaming can potentially be an issue in the absorption column, as well, because it leads to poor process performance;[59] however, foaming can be eliminated effectively by systematic tuning of the surface tension of the CO_2-absorbent system using appropriate ML methods. In addition to these properties, the thermal stability of absorbents, specifically ILs, has been predicted using IL molecular descriptors as input features.[58,60] Thermal stability is an essential property that imposes an upper operating limit on the carbon capture process. By predicting this stability via ML methods, one can minimize the absorbent loss from thermal decomposition, which would be very appealing from an operational-cost viewpoint.

Overall, ML methods have been demonstrated as an attractive alternative to classical thermodynamic approaches (e.g., VLE) for predicting the key properties of various CO_2-absorbent systems, such as equilibrium solubility, with high precision. To develop a tool that is applicable to a wide range of absorbents, the model input features must be tailored to the chemical and physical properties of the absorbent molecule as well as the operational conditions. This approach ensures that the developed model is comprehensive and suitable for predicting the thermodynamic properties of a wide range of molecules that have not yet been considered for carbon capture processes, thus enabling the identification of new alternative molecules for potential future applications.

Process-level optimization

A post-combustion CO_2-capture process based on chemical absorption involves an absorber and a desorber that are connected in a closed-loop fashion. A flue gas with a certain concentration of CO_2 (normally 10%–15%) enters the absorber at the bottom and flows upward, whereas the absorbent (e.g., MEA) solution moves downward, resulting in the absorption of CO_2 by the absorbent. Although the CO_2-lean stream leaves the absorber at the top, the CO_2-rich stream is pumped from the bottom of the absorber to the top of the desorber, where the absorbent is regenerated by heat provided from a reboiler operating with low-pressure steam. The regenerated absorbent (i.e., absorbent-rich stream) is returned to the absorption column for further capture. The product gas at the top of the desorber is cooled in a condenser, and the condensed water is fed back to the desorber, whereas the pure CO_2 gas leaving the condenser is captured. To ensure sufficient contact area in both the absorber and the desorber, trays or packing (random or structured) are used inside the columns.[61] Figure 3 illustrates a simplified CO_2-capture process with an absorber, a desorber, a reboiler, a condenser, and pumps to circulate the solutions.

One of the key challenges facing the current large-scale CO_2-capture plants is the high energy requirement of this process. This challenge highlights the importance of developing a comprehensive tool that can effectively model and subsequently optimize the process to reduce the energy penalty and maximize the CO_2-capture rate. The development of such a tool would also be appealing from an economical viewpoint because it could reduce the cost of capture (i.e., \$/mol CO_2 captured). The traditional mechanistic-based methods of modeling and

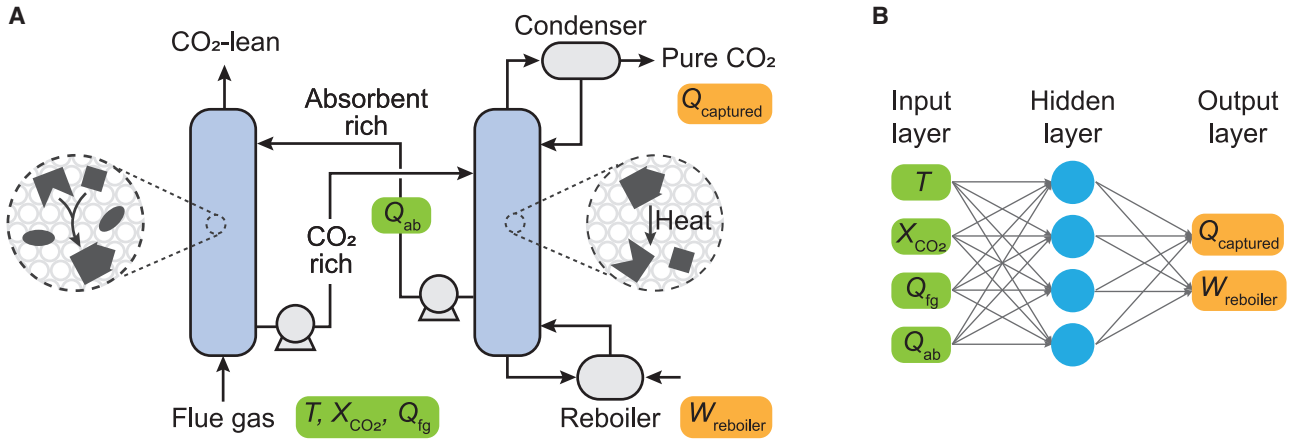

Figure 3. An example of ML implementation for a CO_2-capture process

(A) A simplified flow diagram of an absorbent-based CO_2-capture process with an absorber (left column), a desorber (right column), a reboiler, a condenser, and pumps to circulate the solutions. CO_2 is selectively absorbed from a gas mixture in the absorption column, whereas the absorbent is regenerated by applying heat to the desorber. CO_2 is eventually captured as a pure gas.

(B) An illustration of an ANN as an ML algorithm to correlate the reboiler-specific duty ($W_{reboiler}$) and CO_2-capture rate ($Q_{captured}$) as model outputs to the key operational parameters, including the flue gas temperature (T), CO_2 fraction (XCO_2) and flow rate (Q_{fg}), and the absorbent flow rate (Q_{ab}) as inputs.

optimizing carbon capture processes rely on chemical, fluid-mechanic, and thermodynamic laws, which require extensive knowledge of the underlying physics of the process. The solution for these systems is an iterative procedure accounting for the highly nonlinear nature of the process parameters and outputs, which, for purposes such as certain operating point analysis and optimization studies, is very complex and computationally time consuming.[62,63] In contrast, as a data-driven approach, ML offers a promising alternative that can be developed efficiently with the same level of detail and can be adequately accurate using process data (which are usually readily available), and the evaluation by ML methods is much less computationally demanding than that of traditional methods.

To develop a suitable ML-based model, the input features and output parameters must be selected carefully. The choice must be based upon the objective of the model as well as the physics of the process. In general, key operational parameters, such as the inlet-flue gas temperature, CO_2 fraction, and flow rate, as well as the absorbent-related variables, such as the flow rate, which are widely used as inputs to construct ML models that can predict the essential output parameters, including the process energy requirement and the CO_2-capture rate.[62–66] To train the model, process data are often obtained by performing simulations over various ranges of operational parameter inputs using simulation tools, such as gPROMS.[67] The developed ML model can be used later for implementation in process simulators to perform detailed simulations of larger processes and techno-economic assessments of power plants to minimize capture costs.[68] Figure 3 presents a schematic example in which the operational parameters are the input, and the reboiler-specific duty (which determines the process energy requirements) and the CO_2-capture rate are the output of a ML model.

Adsorption-based carbon capture

Adsorption-based processes offer promising alternative carbon capture technologies. After identifying the source and sink, designing an adsorption-based process involves choosing a solid adsorbent and designing a cyclic process.[69] Clearly, the

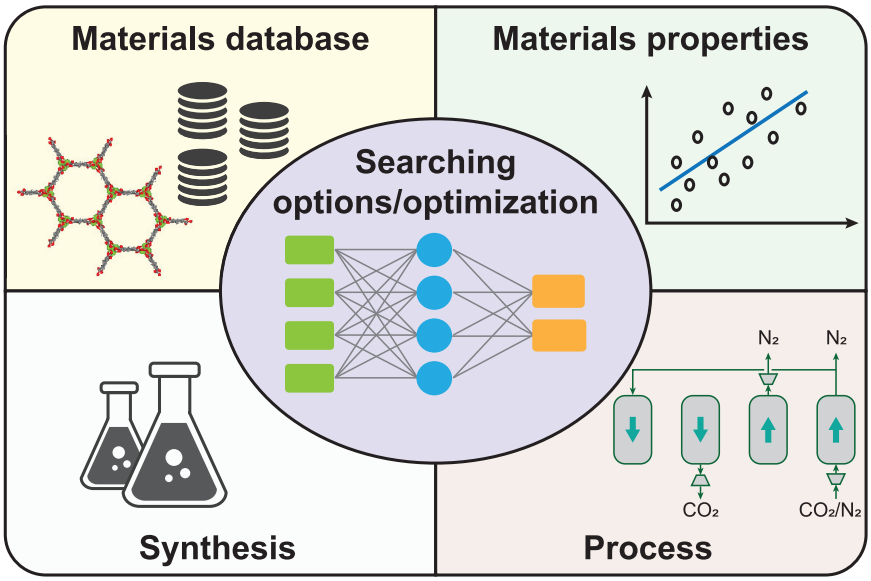

Figure 4. Schematic of ML implementation at various scales in development of adsorption-based carbon capture

From materials to their properties, process-level performance evaluation, and synthesis of the top-performing ones, the challenge of exploring many options must be faced, and ML can help to perform those searches more efficiently.

grand challenge here is to explore a large number of options to find the most cost-effective solution.[70] On one hand, we can choose among a wide range of solid adsorbents, from traditional zeolites and activated carbons to novel classes of porous materials, such as metal-organic frameworks.[71–74] On the other hand, many possible process designs and configurations can be envisioned, e.g., temperature swings, pressure swings, among others. Therefore, development of novel methods for rapid screening and exploring these options is needed. Methods offered by ML and big-data science can facilitate overcoming these challenges.

To systematically design adsorption-based technologies, the several steps[32] sketched in Figure 4 need to be considered. First, the adsorbent material search space, such as MOFs or zeolites, is selected, and the relevant properties, including adsorption properties, thermal properties, etc., are measured or predicted. The next step is to evaluate the materials' performance in the carbon capture processes, where several possible process configurations can be considered. The process configuration includes the choice of cyclic processes as well as how different adsorption columns are configured. This step is followed by an exploration of all these options to find the best combination. Lastly, to realize the full scope of the design, the predicted top-performing materials should be synthesized. ML methods are the natural choice of methodology for solving this type of problem that involves a large number of options because they work best in the limit of large data. In the following sections, we show how ML methods can help us in each step.

Adsorbents and their properties

The research on adsorption-based carbon capture has focused mainly on developing novel adsorbent materials. Conventional porous sorbents, such as zeolites, can possess high selectivity; however, their performance drops dramatically when they are used for flue gases with impurities like water.[75] Therefore, alternative sorbents are needed. Among the different classes of novel materials, MOFs have

received the most attention because of their structural versatility and tunability. MOFs are porous crystalline materials formed by the assembly of metal nodes and organic linkers on a periodic network.[76] The attractive characteristics of MOFs are their extraordinarily high surface area and their chemical tunability, via the choice of metal and functional groups of linkers, to finely control the material properties, e.g., to reach high selectivity and capacity for CO_2 capture. The research on MOFs has led to the development of several material databases containing more than 100,000 experimental-[77–79] and more than 500,000 *in silico*[80,81]-generated structures that can be evaluated for a carbon capture process.[82]

These large databases only constitute a small fraction of the millions of possible MOFs. Therefore, of fundamental importance is knowing whether these structures cover the actual design space of relevant MOFs for carbon capture applications. This question can be answered using unsupervised ML methods with attempts to make sense of unlabeled data to uncover the underlying patterns and distributions in those data. For that analysis, the first step is to map structures onto numbers to quantify their similarities. The design space of MOFs for carbon capture applications comprises pore geometry and topology and the chemistry of metal nodes, linkers, and functional groups. Moosavi et al.[83] adopted graph-theoretic descriptors for each of those characteristics to analyze the structural diversity in MOF databases. Although it is desirable to screen or analyze a database that has sufficient diversity to represent all possible structures, this study showed that each database has a significantly different distribution and systematic bias toward specific types of chemistries and geometries, e.g., the hypothetical databases severely biased toward a few metal types. Notably, it was shown that this lack of diversity has led to wrong conclusions in the past. Interestingly, using this approach, it can be determined whether a new material adds information to the currently available structures or has only a small variation of existing structures.

The next step is to be able to predict specific material properties for these structures. Clearly, the countless number of structures make any brute-force, experimental or computational screening infeasible. ML methods can help to efficiently predict these material properties and filter out those structures with poor performance. Essential in these studies is featurization, in which, a material is represented with a set of descriptors. In particular, for the adsorption properties, both pore geometry (i.e., characteristics of pore shape and size) and the chemistry of materials are significant. This has motivated development of several representation approaches that are discussed in detailed in the literature.[31] Among them, for geometric representation, simple geometric descriptors, such as pore size, surface area, pore volume, etc., are found to be effective. In addition, persistent homology from topological data analysis was introduced as a method for assigning similarity among the shapes of pores.[84–86] For the chemistry of materials, revised autocorrelation descriptors[83] and the descriptors of atomic-property-weighted, radial-distribution functions[87,88] were shown to be effective in ML models for predicting several adsorption-related properties of nanoporous materials. In addition, caution is required when using the ML models to predict the properties of new materials. Because ML methods do not use the underlying physical laws for new predictions, it is crucially important to ensure the model is used within its domain of applicability.[89,90] For that purpose, the use of uncertainty metrics to assess the reliability of the predictions is important. In addition, it was shown that training models using smaller, more diverse datasets lead to more transferable ML models for prediction of the properties for new materials.[83,91]

These studies were focused on the prediction of adsorption properties at a specific pressure, temperature, and gas composition. However, for evaluation of materials in process optimization, a typical need is to have access to the full isotherm and mixture-equilibria data. Sun et al.[92] developed a deep neural network model, called SorbNet, trained on simulation data to predict full isotherm, mixture, and equilibrium properties. Using that approach, they successfully obtained a continuous isotherm function that was used to optimize a chemical process.

Performance evaluation

Simple performance indicators (PIs) based on material properties, such as working capacity, selectivity, and heat of adsorption, were used in many studies aiming to design better adsorbents for CO_2-capture processes.[71–73] Although these basic PIs allowed for a preliminary assessment of the performance, it is now well known that these metrics do not represent our ultimate goal, which is a reduction in the cost of the capture process; hence, there are several recent studies that incorporate process modeling and optimization for performance evaluation.[93–95] An excellent example is synergistic design of materials and adsorption processes.[70] A comprehensive review on this topic can be found in the literature.[69]

Cyclic adsorption processes incorporate various operational strategies for gas separation, including temperature-swing adsorption (TSA), pressure/vacuum-swing adsorption (PSA/VSA), temperature-vacuum swing adsorption (TVSA), steam regeneration, etc. Each of these processes can be designed by combining a variety of different cycle configurations. For example, in a PSA process, with a combination of cycles, such as adsorption, blow down, regeneration, etc., a wide range of adsorption processes can be designed. In the modeling of such a cyclic process, solutions are needed for a system of partial differential equations (PDEs) governing the mass, momentum, and heat transport phenomena, depending on the model details.[96,97] Obviously, for even one material, finding the optimal process design is not straightforward because a large range of options can be considered. Several recent studies have addressed this challenge using ML methods. For example, Subraveti et al.[98] developed surrogate-assisted optimization (SOpt) to directly predict the cycle steady state to reduce the cost of solving systems of PDEs. The accuracy and reliability of the model was demonstrated on a complex eight-step PSA process, which is encouraging for future incorporation of ML methods for process modeling and optimization.

Exploring the options

The final step in evaluating adsorption process options is to search for the optimal material-process combination. Evidently, we cannot exhaustively search all the possible options and perform brute-force calculations on all materials and process. A pragmatic approach is to restrict the search to the materials that have already been synthesized.[99] In one of the first and recent efforts along this line, Burns et al.[100] linked molecular simulation and process modeling to evaluate more than 1,600 MOFs for post-combustion CO_2 capture using a fixed VSA process. Among those MOFs, only ~500 materials met the 90% purity and 95% recovery requirements set by the U.S. Department of Energy. To assist that search, they trained an ML model to rapidly evaluate which materials would meet those purity and recovery requirements, with an accuracy greater than 90% in identifying the promising materials.

A complementary approach is to use the data from those screening studies to extract design rules. In particular, such information can provide valuable guidelines for

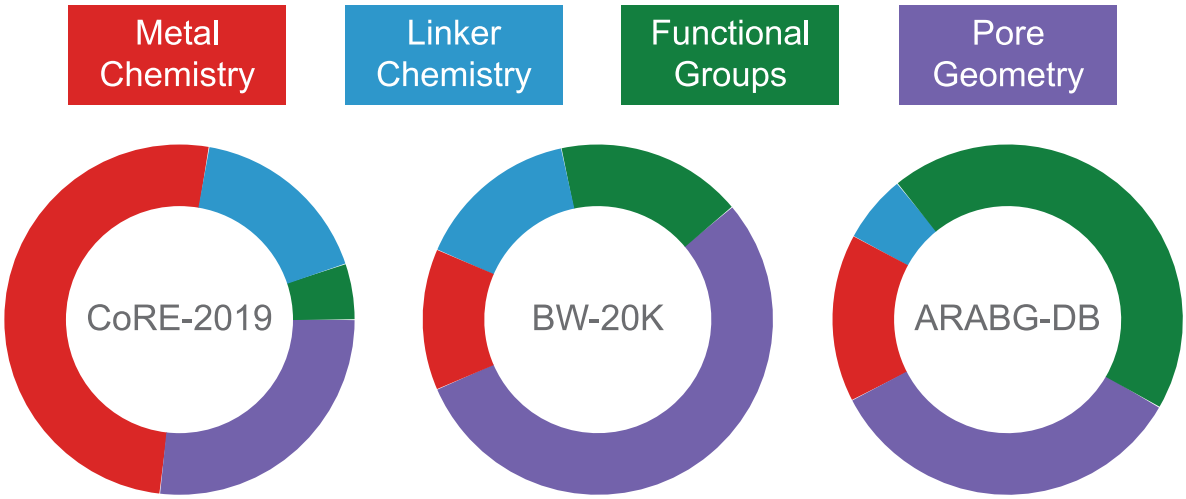

Figure 5. The relative importance of material characteristics for CO_2 adsorption

Pie charts showing the relative importance of variables extracted for three MOF databases, including experimental MOFs (CoRE-2019)[79] and two hypothetical databases (namely BW-20K and ARABG-DB),[23,101] with different structural diversity. Biased distribution in materials leads to different importance in material characteristics. The figure was adapted from Moosavi et al.,[83] published by Springer Nature under CC-BY license.

experimental researchers seeking to design and synthesize next-generation high-performance MOFs. Although exploratory data analysis can be used to extract structure-property relationships, ML methods are most effective for extracting these rules from high-dimensional data. For the design of materials for carbon capture, it is fundamentally important to develop an understanding of how changes in the material chemistry (i.e., change of metals, linkers, or functional groups) or pore geometry of MOFs affect CO_2 adsorption properties. In a recent study with several MOF databases, the relative effect of material characteristics on the CO_2 uptake at low pressures was analyzed.[83,101] Noticeably, it was found that, for different databases, different relative importance can be ascribed to each of these properties (Figure 5).[83] Careful inspection of these results shows that, depending on the diversity of the databases, the conclusions might not be generalizable to the entire chemistry of a material class. This points to the significant importance of diversity in material databases, which needs to be considered when drawing conclusions from ML models.

The ultimate goal of these studies is to design the best carbon capture setup. Generative ML methods can be used to generate new materials and process combinations, providing direct optimization for the final goal. These methods have only recently been applied for the design of porous materials and, in particular, for the discovery of materials with target properties. Kim et al.[102] developed a generative adversarial network (GAN) to generate new zeolite structures with user-desired properties, in this case, heat of adsorption for methane. Another promising technique, one that uses variational autoencoders to allow continuous optimization of material properties, was recently applied to the design of MOFs.[103] Further development of these approaches can greatly benefit the field of carbon capture materials and technology.

Lastly, ML models can be recruited to accelerate synthesis of the discovered materials. To realize the final aim of material-process design, it is necessary to synthesize the discovered promising candidates. To synthesize new materials, chemists often use chemical intuition to explore a large number of options, such as choice of reactants, reaction conditions, etc. Data-driven methods can be used to extract that chemical intuition from a dataset of failed and successful experiments and to apply

the results toward developing more-efficient material-synthesis planning. For porous materials, initial efforts are encouraging; this approach has been successfully demonstrated, for instance, in the evolution of optimal synthesis conditions for the synthesis of high-quality MOFs.[104] In addition, ML has been used to find structure-directing agents for the synthesis of zeolites.[105]

FUTURE OPPORTUNITIES FOR ML IN CARBON CAPTURE

As an emerging tool, ML has been shown to be an effective method with great potential for accelerating advances on many fronts in carbon capture technology. Here, we name some of the future directions in which ML deployment could benefit the field.

Absorption-based technologies

Many aspects of absorption-based carbon capture processes, ranging from the molecular to the process level, have benefited from ML implementation, providing researchers with a reliable tool that can predict the outcome of a particular system or configuration without the need for experiments. Considering its demonstrated effectiveness in carbon capture processes, together with the rapid progress in the field of ML, many unexplored areas of this process are expected to benefit from ML tools in future investigations. For example, a key concern in thermal processes is amine degradation, which could be studied using ML methods because numerous structured datasets on the degradation of various amines over a wide range of operational conditions can be extracted from the literature. By developing a reliable and comprehensive model, the degradation rates and products as a function of the operational conditions can be predicted, which could provide guidelines for selecting the upper boundaries of the process to minimize amine loss. As another example, ML can be used to predict the absorbent solubility in a solvent (as opposed to the CO_2 solubility in the absorbent). This absorbent solubility is a key parameter because higher solubility results in greater absorption capacity for CO_2. These are only a couple of examples of the many future opportunities for ML to assist in absorbent-based carbon capture processes at a molecular scale.

In terms of the process-level optimization, reinforcement learning can be considered to obtain the optimal operational conditions using hybrid information from both process simulation and real-plant data. In reinforcement learning, the machine is exposed to an environment in which it trains itself continually using the trial-and-error method, thereby learning from experience to make accurate decisions for the future.[106] The optimal operational conditions obtained by the conventional, supervised ML models, trained solely by process simulation data are likely restricted to the simulated model selected. However, implementation of reinforcement learning techniques incorporates the real-plant data to update the learning scheme and ultimately reach more-accurate and representative optimal operational conditions. This is an area that has great potential for future carbon capture process development in which ML is used in conjunction with process simulation and real-plant data.

Adsorption-based technologies

Although most of the research efforts so far on designing new materials have focused on improving the CO_2 and N_2 adsorption properties, there are several other material properties that are now seen to be bottlenecks for deployment of adsorption-based carbon capture technologies. To name a few, the role of impurities, such as that in water,[107] on adsorption properties, as well as recyclability of materials that are dominated by chemical, thermal, and mechanical stabilities, need to be

thoroughly investigated.[108,109] In particular, because accurate prediction of these properties is computationally demanding, the development of ML methods can enable large-scale screening and fast prediction of these properties. In addition, although our eventual aim is to explore the full design space of materials using hypothetical structures, we face the issue of synthesizability of the discovered structures. The underlying challenge is to be able to predict whether a material can be synthesized. The development of ML models to assess synthetic feasibility and incorporation of such a metric to bias search algorithms can have a large effect on material development for carbon capture technologies. The recent article by Boyd et al.[23] addresses some of these challenges by incorporating the chemical intuition of chemists into material design. To do so, after a large-scale screening of MOFs to identify good adsorption sites for CO_2, they developed a secondary, smaller library of hypothetical materials for which they have indications of synthetic feasibility in addition to low cost and high chemical and thermal stability.

The development of specific ML methods and algorithms to perform learning tasks more efficiently will be an important topic of research. Particularly, recent developments in deep learning, for example, multi-task learning and transfer learning,[110] could facilitate the learning procedure and help to predict material properties with high accuracy when we only have access to limited data. The underlying methodology used in these approaches is to improve the learning by transferring parameters or features as well as sharing contextual information.

For process modeling and optimization, applications of ML have just started. For a specific process design, e.g., a four-step PSA process with defined cycles, the continuous nature of the problem allows for successful application of ML methods for rapid evaluation of different process configurations. In particular, further developments are needed in developing physics-informed ML methods for adsorption, for example, by developing models that are compatible with the physics of heat and mass transport. In addition, versatile ML models and architectures that can incorporate the modular design of adsorption processes would allow screening of a wide range of processes for a specific material. Further scientific insight in the form of structure-performance relationships can enable a rational design of adsorption-based processes.

The sheer complexity of the multi-stage design of adsorption-based technologies that considers materials and processes, inevitably drives the requirement for new solution techniques. Indeed, this topic can greatly benefit from, and potentially be revolutionized by, using ML methods to make it feasible to explore a wide range of options efficiently. Several exciting and broad directions should be developed and researched, including methods for careful assessment of diversity and bias, and methods for assessing uncertainty and error in ML predictions and how those errors propagate through the full design process, as well as algorithms for multi-objective searches.

Lastly, most of the current investigations have targeted the development of ML models for carbon capture under pre- and post-combustion conditions. Other carbon capture applications can equally benefit from these ML methods; among which, direct air capture using MOFs has gained enormous attention despite controversy about the cost-effectiveness of such technology.[111–114] ML methods can be used to evaluate the feasibility of the technology as well as guide the future discovery and synthesis of high-performance MOFs for air-capture purposes.

Other carbon capture technologies

Several other approaches have been widely investigated for carbon capture, including membrane-based[115–117] and electrochemical[118–120] systems. These approaches require further development on many fronts before they can be considered for large-scale implementations. As part of that development, ML methods may have a crucial role in effectively improving these processes.

Membrane-based technology has recently received substantial attention, and researchers have aimed to develop a modular system that can selectively remove CO_2 from different sources. The primary characteristics of the membrane, including the selectivity, durability, and cost, can be correlated to the structural properties of the membrane backbone through a well-trained ML method. Such correlations can help in clarifying the effect of these properties on the membrane characteristics and, subsequently, enable the fabrication of membranes that are highly selective to CO_2, durable over long-term operations, and cost effective. As a recent example, an ML-based model was trained using a topological, path-based hash of the polymer-repeating unit of the membrane, with the goal of discovering new materials optimized for separating a given gas pair. To test the model accuracy, two of the most promising polymer membranes predicted by this approach were synthesized, and it was found that the experimental CO_2 separation performance was very close to that predicted by the model.[121] This approach can ultimately pave the way for effective implementation of membrane-based technology for large-scale carbon capture operations.

Electrochemical systems designed for carbon capture are emerging as an alternative approach that can potentially separate CO_2 through reversible cycles with energy requirements comparable to those of state-of-the-art thermal processes.[122–124] Because of their electrical nature, renewable sources of energy, such as solar and wind, could be used to compensate for the energy penalty of such processes, offering an attractive route for CO_2 capture with a minimal carbon footprint. Several electrochemical approaches have been introduced for carbon capture from both point sources (such as industrial flue gas)[125–127] and dilute streams (e.g., air)[128,129]; however, these systems are in the early stages of development. Along with experimental and fundamental modeling studies, ML can further advance that process. That advancement could be achieved through various means, including the development of absorbent systems, electrodes, and cell configurations, as well as studies on stability, energy penalties, and CO_2-capture rates. Because of the similarity between these technologies and well-established electrochemical energy storage systems (such as batteries), the expertise developed in implementing ML methods for batteries[130–132] can provide a roadmap for employing these approaches for electrochemical CO_2-capture systems.

CONCLUSIONS AND OUTLOOK

In this perspective, we reviewed ML implementations for both absorbent- and adsorbent-based carbon capture processes. We discussed how ML has emerged as a useful tool to predict the thermodynamic properties of CO_2, especially the CO_2 solubilities of various absorbents. In that regard, we discussed the prediction of CO_2 solubilities for both conventional amines used in state-of-the-art carbon capture power plants and emerging alternative absorbents with great potential for future carbon capture processes, such as ILs. The prediction of other thermodynamic properties, such as the viscosity and density, using ML was also discussed. ML algorithms have further been used at the process level to model entire carbon capture plants,

with the aim of identifying optimum operational conditions to obtain the minimum specific steam duty and maximum CO_2-capture rate. In the context of adsorbent-based carbon capture, MOFs featuring the advantages of high selectivity and capacity have benefited from ML, enabling a down-selection from large numbers of potential candidates and obtaining the information and guidance required to accelerate the discovery of novel advanced materials. Large databases of different MOFs can be made computationally by tuning chemistry- and topology-related properties; however, finding an appropriate high-performance material in such a large database through experiments or traditional theoretical calculations is undoubtedly challenging. This task can be greatly simplified by the careful implementation of appropriate ML algorithms.

For a data-driven tool, such as ML, data acquisition is highly important for developing a model that can effectively reflect the underlying nonlinearities, complexities, and intricacies of the system parameters. As mentioned earlier, the data used to train ML models are obtained from either experimental measurements or computational simulations. The ranges and conditions under which the data are generated are critical for developing a comprehensive model; hence, the ranges and conditions must be carefully established to represent the actual process. For example, several datasets on CO_2 solubility have been obtained by experimental measurements in which various CO_2 partial pressures (mixed with N_2) were purged at different temperatures, and the solubility values were measured. A model developed upon this dataset is valid as long as it is used to predict the solubility values when the absorbent is exposed to the same binary gas composition (i.e., CO_2 and N_2). However, the model likely fails to accurately predict the solubility values when the absorbent is considered for different applications, e.g., direct air capture, in which a major quantity of O_2 is present. Therefore, the experimental (or simulation) ranges and conditions must be tailored to those of the actual application.

Despite the high accuracy of the ML models developed for various applications in carbon capture, their limitations should not be ignored. Unlike traditional theoretical calculations, the developed ML models rely solely on data and are not directly derived from thermodynamic principles. This aspect highlights the importance of the features used in training ML models; these features must carry physical insights to be sufficiently representative of the system's underlying thermodynamic fundamentals. Thus, this method requires the systematic development of descriptors that are neither too general nor too detailed. Group contributions and chemistry/topology descriptors are two good examples used for ILs and MOFs, respectively, as elaborated earlier. Overall, ML is a powerful tool that has shown great potential for affecting various aspects of carbon capture technologies, if carefully implemented. Well-developed ML tools can provide valuable guidelines for experimental researchers and computational chemists to rapidly advance smart carbon capture technologies as a necessary step toward climate-change mitigation.

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AUTHOR CONTRIBUTIONS

All authors contributed to writing and revising the manuscript.

DECLARATION OF INTERESTS

The authors declare no competing interests.

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