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Data-Driven Design and Discovery of Metal–Organic Framework/Polymer Mixed Matrix Membranes

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ABSTRACT

Metal–organic frameworks (MOFs) are one of the most promising classes of porous materials thanks to their tunable chemistry, structural diversity, and a variety of useful structural properties such as very high porosities and extraordinarily large surface areas. Mixed-matrix membranes (MMMs) that incorporate MOF fillers into polymers have emerged to overcome the selectivity-permeability trade-offs inherent to traditional polymer membranes for gas separations. MOF/polymer MMMs are currently at the forefront of membrane research thanks to their ability to bridge the performance gaps of pure polymers and the practical limitations of pristine MOFs. While experimental studies have demonstrated the strong benefit of using of MOF/polymer MMMs for many different types of separations, the rational design of novel MMMs remains a complex, multi-scale challenge. Integration of machine learning (ML) to current experimental and computational studies will be central to unlocking the industrial potential of MOF/polymer MMMs by guiding materials selection, predicting membrane performance, and even synthesis conditions. This perspective explores how ML is reshaping the design and discovery of MOF/polymer MMMs by discussing current progress, opportunities, and challenges of uniting computational innovation with experimental validation to create the next generation of MOF-based MMMs.

1 | Introduction

Gas separation membranes are a critical part of the industrial processes, offering energy-efficient alternatives to traditional cryogenic or adsorption-based separations [1]. While polymer membranes lead the market because of their low cost, scalability, and well-known fabrications, they are intrinsically limited by the permeability-selectivity trade-off described by the Robeson upper bound [2–4]. Metal–organic frameworks (MOFs), with their tunable pore structures and chemical functionalities, have the potential to break through this trade-off by providing size- and affinity-based selectivity for target gas molecules. MOFs are crystalline frameworks in which metal nodes are interconnected by organic linkers, forming highly porous structures with virtually limitless chemical and topological diversity. Such chemical tunability has rendered MOFs highly attractive materials for various chemical applications, and among them, gas separation has been the most widely studied and, hence, currently the most mature field [5–7].

MOF-incorporated polymer membranes are known as MOF/polymer mixed matrix membranes (MMMs). They combine mechanical, chemical robustness, and easy processability of polymers with tunability and variety of MOFs, creating hybrid materials capable of surpassing the performance of either component alone. A very large number of studies reported synthesis, characterization, and separation performances of MOF/polymer MMMs, and several excellent reviews have addressed both the opportunities and the challenges of using MMMs [8–13]. Experimental and computational studies

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have shown that using MOFs as fillers in polymer membranes increases permeability and/or selectivity of the pristine polymer [14, 15]. For example, molecular simulations performed over >180,000 different types of MOF/polymer MMMs for 11 gas separations including CO_2, CH_4, N_2, H_2, O_2, and He gases showed that incorporating MOFs into polymers enhances the permeability of nearly all gases across most polymers as represented with the red regions in Figure 1, thanks to the very highly porous structures of MOFs [16]. On the other hand, MOF fillers do not substantially change the polymer membranes' selectivities, as indicated by the large yellow regions. In some cases, MOF fillers reduce the selectivity of certain polymers, although the extent of this decrease is typically negligible. Nevertheless, many MOF/polymer MMMs were shown to surpass the upper bound for different gas separations through improvements in gas permeability, highlighting the potential of MOF/polymer MMMs for industrial applications.

However, designing high-performance MOF/polymer MMMs is far from straightforward. Performance of these membranes, mainly quantified by gas permeability and selectivity, depends not only on the inherent properties of MOF and polymer but also on the often-complex interfacial interactions between filler and polymer [17–19]. Therefore, the design of MOF/polymer MMMs demands careful selection of both the MOF and the polymer components. Given the vast and continually expanding library of MOFs, identifying the most appropriate filler for a specific polymer is a critical challenge, and relying solely on experimental methods to screen hundreds of thousands of available MOF candidates is impractical. Computational studies, more specifically, molecular simulations, have become a powerful tool for identifying the most effective MOF fillers for MMMs, allowing researchers to systematically screen diverse MOF/polymer combinations, to predict their gas adsorption and transport properties, and to investigate their interfacial structures [20]. These computational studies typically combine two types of molecular simulations; grand canonical Monte Carlo (GCMC) to assess gas adsorption isotherms and molecular dynamics (MD) to compute gas diffusivities in MOFs. This molecular-level gas transport data is used to assess the gas permeability of MOFs and, combined with the gas permeability of polymers, generally available in the literature thanks to the widespread investigation of polymers as membranes, via continuum permeation models [21] such as Maxwell model to calculate permeability and selectivity of the MMMs [22].

While this approach can accurately reproduce experimental gas permeability data of many MOF/polymer MMMs, its applicability is still constrained by the extensive computational time and resources required. Performing MD simulations for a very large number of MOFs and multiple gas types requires collecting and analyzing several independent trajectories of adsorbed molecules within the MOFs. Furthermore, this approach does not consider the MOF-polymer interfacial effects, which are critical for the performance of MMMs [23, 24]. Molecular simulations supported by quantum chemical calculations are used to capture structural relaxation and void formation at the interface, but these calculations are generally limited to a single type of MOF and polymer due to the excessive time requirement of the calculations [25, 26]. Even when all these simulations and calculations are completed, extracting structure-performance relations from the resulting large datasets remains a significant challenge without the aid of data science tools.

As in many disciplines, machine learning (ML) has become increasingly important in the fields of membrane design and MOF research, offering a powerful alternative to traditional and resource-demanding methods. ML has been recently used for a large variety of membrane-based applications, such as the identification of the most promising membrane candidates for H_2S/CH_4 separation [27], investigating the fundamental steps of thin-film formation of polymers [28], optimizing the forward osmosis membrane efficiency for water treatment [29], and predicting the solute rejection using an open-access membrane dataset [30]. In the field of MOFs, ML models trained on high-quality computational or experimental data of MOFs have been shown to accurately predict the key properties of gas molecules in MOFs, such as uptakes and diffusivities, in addition to predicting other critical metrics of MOFs, such as stability at a fraction of the computational cost [31]. Therefore, ML enabled rapid exploration of vast MOF databases, accelerating the discovery of materials with optimal performance for different gas separations.

As shown in Figure 2, the number of studies focusing on MOF/polymer MMMs through experimental and computational techniques has been steadily increasing, underscoring the growing need for ML to efficiently analyze this expanding body of data. ML can be used to screen large numbers of MOF-polymer combinations for materials selection, predicting the optimum synthesis conditions of MOF/polymer MMMs, and assessing these membranes' performances. Applying ML for examining MOF/polymer MMMs has recently started, and the number of works is growing very rapidly. Motivated from this point, in this perspective, we first reviewed the current state-of-the-art in the field and then focused on the potential of ML for enabling the rational design and discovery of MOF/polymer MMMs by uncovering complex relationships between structural descriptors of MOFs and polymers, and their impacts on the MMMs' performances. Since we only focused on the impact of ML on the design and discovery of MOF/polymer MMMs, we kindly direct the readers to several existing reviews [31–33] on the basics and implementation of ML, which covers different algorithms and technical details.

2 | Result and Discussion: State-of-the-Art ML Approaches for MOF/Polymer MMMs

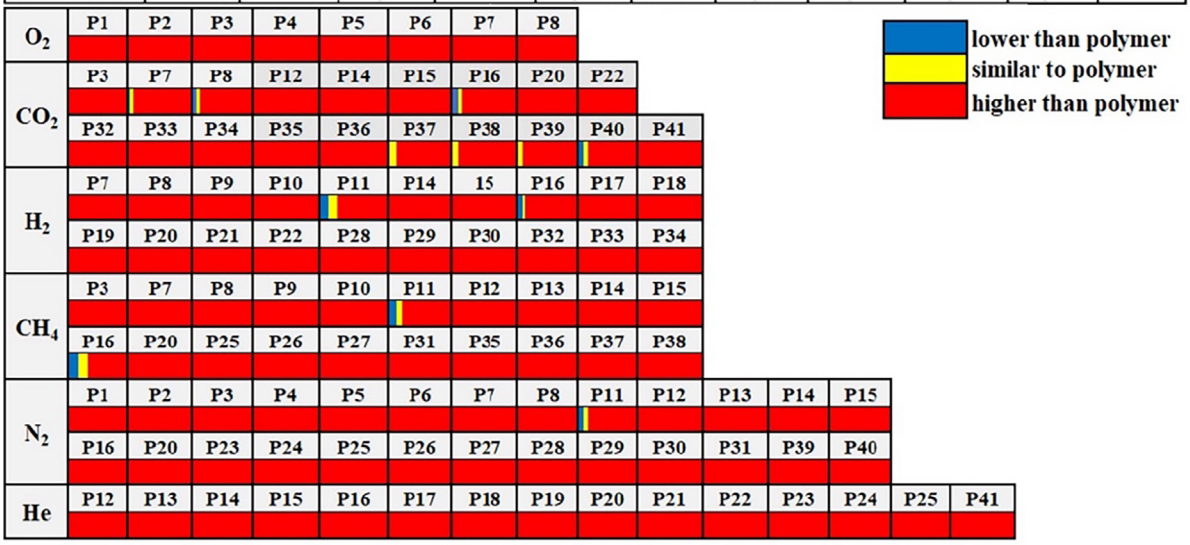
The first study [34] that used ML to evaluate gas separation performance of MMMs examined 31,494 distinct types of MOF/polymer MMMs, a very large material scale that is infeasible to explore experimentally. A recently compiled database of synthesized MOFs was used, and fillers were selected based on their pore sizes for the effective adsorption and transport of gas molecules. Uptakes and diffusivities of He, H_2, CH_4, and N_2 gases in 5,249 MOFs were computed by GCMC and MD simulations. ML models were then trained on this data to predict gas adsorption and diffusion properties of MOFs, leading to R^2 values between 0.96–0.99 and 0.83–0.97, respectively. These ML models used 20 different descriptors of MOFs; structural features related to pore

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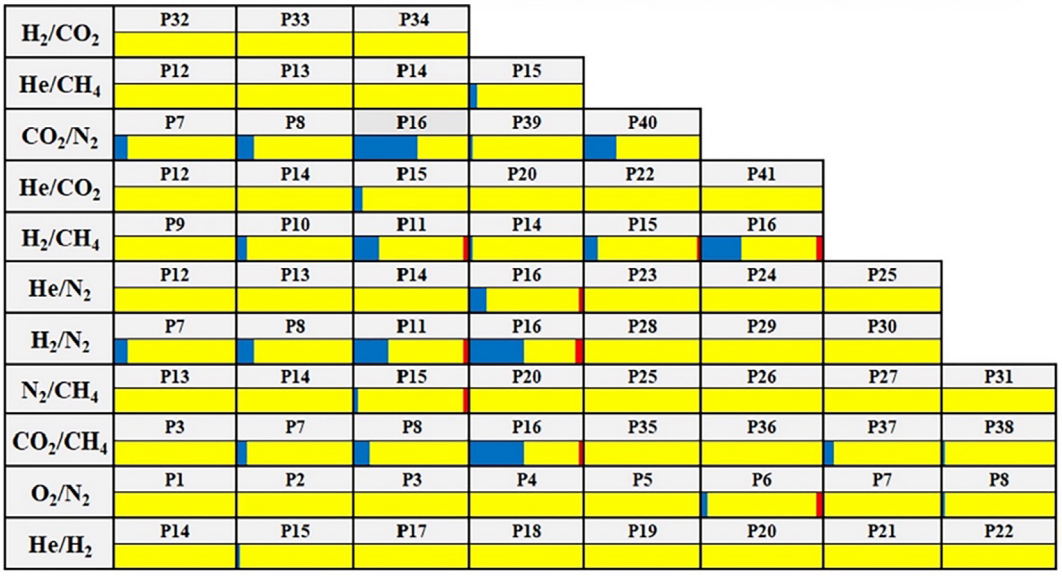
(a)

	H_2/CO_2	He/CH_4	CO_2/N_2	He/CO_2	H_2/CH_4	He/N_2	H_2/N_2	N_2/CH_4	CO_2/CH_4	O_2/N_2	He/H_2
# of MOF	1194	864	2124	314	3255	864	3255	5599	2124	5599	846
# of Polymer	3	4	5	6	6	7	7	8	8	8	8
# of MMM	3582	3456	10,620	1884	19,530	6048	22,785	44,792	16,992	44,792	6768

(b)



(c)



P1: BPDA-ODA	**P12**: 6FDA-PMDA(10/90)-TAB	**P23:** 6FDA-6FpDA:DABA(2:1)	**P34:** 1,1-6FDA-DIA
P2: Polyetherimide	**P13**: 6FDA-TAB	**P24:** TMHFBPA-I/T	**P35:** PVSH doped polyaniline
P3: 6FDA/PMDA(25/75)-TAB	**P14**: Hyflon AD60X	**P25:** Hyflon AD	**P36:** TADATO/DSDA(1/1)-DDBT
P4: Sulfonated brominated PPO	**P15**: Teflon AF-2400	**P26:** 6FDA-mPDA	**P37:** PI-5
P5: BADBSBF-BTDA	**P16**: PTMSP	**P27:** PTMSP-co(60/40)	**P38:** PDPA-3f
P6: PTMSDPA	**P17**: Viton E60	**P28**: PBOI-2-Cu $^{+}$	**P39:** MEEP
P7: PIM-7	**P18:** Nafion 117	**P29**: NTDA-BAPHFDS(H)	**P40:** PTMGP
P8: PIM-1	**P19:** Viton GF	**P30**: PAI-3a	**P41:** Hyflon AD60
P9: DAPHFDS(H)	**P20:** Cytop	**P31**: Teflon AF-1600	
P10: 6FDA-DDBT	**P21:** Fluorinated Polynorbornene	**P32:** Liquid polyester (HBA/HNA-30/70)	
P11: PTMSP-co(95/5)	**P22:** Hyflon AD80	**P33:** Polyaniline (redoped)	

FIGURE 1 | (a) Numbers represent MOFs, polymers, and MOF/polymer MMMs examined for gas separations. The effect of using MOF fillers on gas permeabilities (b) and selectivities (c) of polymers are shown with colors. Red (blue) represents that MOF/polymer MMMs exhibit higher (lower) values compared to polymer. MOF/polymer MMMs having similar performances (±10%) with pristine polymers are shown with yellow. Adapted with permission [16]. Copyright 2022, Elsevier.

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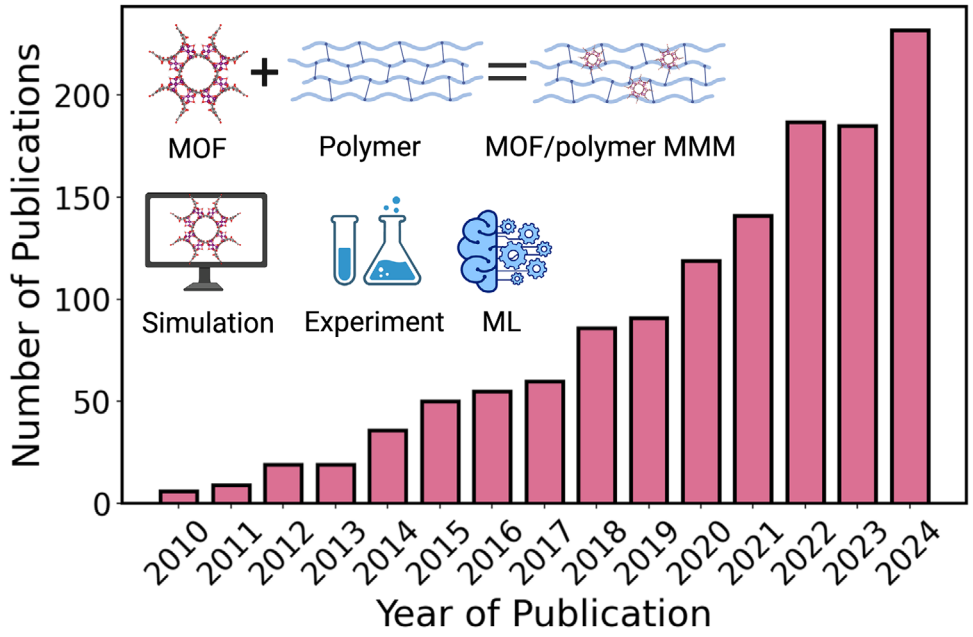


FIGURE 2 | The number of MOF/polymer MMM publications reported in the literature obtained through Scopus database featuring the terms "metal organic framework" (or MOF) and "mixed matrix membranes" (or MMM) in the article title, abstract, or keywords. Accessed on August 25, 2025.

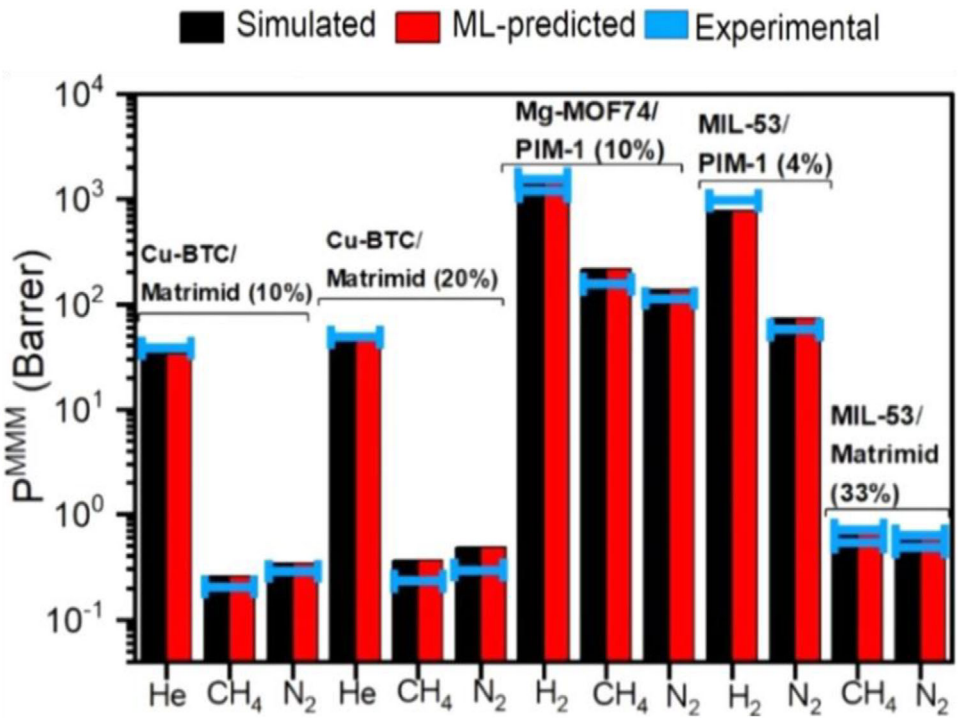


FIGURE 3 | Comparison of ML predictions, simulation results, and experimentally measured gas permeabilities for different types of MOF/polymer MMMs. Adapted under the terms of the CC BY 4.0 license [34]. Copyright 2022, ACS.

size and pore geometry, in addition to chemical and energetic features, such as elemental analysis and heat of adsorption values. Gas permeabilities of MOFs obtained from the ML models were used together with the polymers' permeability data collected from the experimental literature to estimate He, H_2, CH_4, and N_2 permeability of MOF/polymer MMMs. As shown in Figure 3, the comparison of ML predictions with experimentally fabricated CuBTC/Matrimid, Mg-MOF-74/PIM-1, MIL-53/Matrimid, and MIL-53/PIM-1 MMMs demonstrated the potential of ML for rapid and accurate prediction of permeabilities in MOF/polymer MMMs, effectively bypassing computationally demanding molecular simulations.

Given the extensive experimental effort required to fabricate and characterize even a single MOF/polymer MMM, from material selection to membrane fabrication and performance measure-

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ments, the ability of ML to provide accurate estimates across such a vast design space is valuable and transformative for accelerating membrane material discovery. However, Daglar's work used ML to predict only the pristine MOFs' gas permeability data, not the MOF/polymer MMMs' permeability. Developing ML models that use individual features of polymers and MOFs to make predictions directly for MOF/polymer MMMs appeared later, as will be discussed.

The next pioneering study [35] in this field used ML to experimentally design a new MOF/polymer MMM for CO_2 capture. Experimental data reported in the literature for CO_2/CH_4 separation performances of MOF/polymer MMMs, involving 36 MOF and 41 polymer types, were collected and used to train Random Forest ML models. These ML models used not only MOFs but also MMMs' features, such as MOF loading and thickness, as inputs to predict the relative CO_2 permeability, which was defined as the ratio of permeability of MMM over that of the control polymer. ML models indicated that MOFs with certain pore sizes and surface areas are promising for MMM applications. These insights guided the selection of two different types of MOFs with desired characteristics, which were subsequently synthesized and used to experimentally fabricate MMMs with Pebax and PIM-1 polymers. The CO_2 permeability measured in the experiments agreed well with the ML models' prediction, while relatively more differences were observed in the relative CO_2/CH_4 selectivity. The resulting MMMs surpassed the Robeson's upper bound, indicating that using ML does not only provides some estimates about gas permeability but also it can guide the experimental design of high-performance MMMs.

A similar approach was used to investigate the CO_2/N_2 separation performance of MOF/polymer MMMs by developing a genetic algorithm-optimized artificial neural network model [36]. The authors compiled all available data on CO_2/N_2 separation performance of MOF/polymer MMMs reported in journal publications, resulting in a dataset covering 43 distinct MOFs and 19 different polymers. The study placed particular emphasis on feature engineering by employing target encoding to represent both MOF and polymer types. MOF-related properties such as particle size, surface area, pore diameter, and MOF loading in the membrane were considered in addition to molecular descriptors representing polymers' chemical characteristics. The ML analysis revealed that the most influential features for predicting the gas permeability of MOF/polymer MMMs were MOFs' surface area and particle size, whereas the most critical features affecting selectivity of the MMMs were type of polymer and MOF. As anticipated, these ML results emphasize that the intrinsic properties of both MOFs and polymers are crucial for shaping the separation performance of MMMs.

The last two studies we reviewed compiled MMM data from the literature. Since similar types of MOFs and polymers were used in the experimental fabrication of MMMs, the collected dataset was limited to less than 50 different MOFs in those two studies. Consequently, the ML models were trained on a very restricted number and type of MOFs, limiting their ability to capture the full diversity of MOF chemistry. To account for the large structural and chemical variety of MOFs, performing molecular simulations represents a valuable strategy. For example, molecular simulations were combined with ML to assess the CO_2 separation performance of 54,117 different types of MMMs, which captures 9 polymers and 6,013 MOFs [37]. GCMC and MD simulations were first carried out to calculate the gas adsorption capacities and diffusion coefficients of MOFs. The resulting data were then integrated with experimentally reported polymer gas permeability values through the Maxwell model to predict the separation performance of MOF/polymer MMMs. ML models were developed to estimate not the permeability but the TSP (ln of permeability multiplied by the selectivity) values of the MMMs when the structural properties of MOFs and polymers are given. The Shapley analysis, showing the impact of feature variables on target data in ML models, identified the limiting diameter and the largest diameter of pores as the most critical MOF features whereas the fractional free volume and density were shown to be the most vital polymer features. Here, the ML models were trained using only these two properties of polymers so the effect of other structural and chemical properties of polymers were unknown.

Using a similar computational strategy, a recent work [38] combined high-throughput computational screening of MOFs with the experimentally reported data of polymers to generate a very large dataset of 456,872 different types of MOF/polymer MMMs for He/CH_4 separation. As represented in Figure 4, GCMC and MD simulations were performed for 10,143 MOFs and combined with the experimental data of 26 polymers through the Maxwell model. The ML models were trained to directly predict the performances of MOF/polymer MMMs, where the target variables were the MMMs' He permeability and selectivity, and the input variables were the polymers' permeability and selectivity in addition to structural properties of MOFs. The Shapley analysis was performed to get data-driven insights from the ML models about the selection of materials. Results revealed that the polymers having He permeability >1600 Barrer and He/CH_4 selectivity >900 must be selected and combined with MOF fillers having pore sizes between 2.6 and 3.2 Å to obtain high-performance MMMs. In ML models, polymers' gas permeability and selectivity, already known to be the key factors to significantly influence MMM performance through Maxwell models, were used as input features. While this is a valuable starting point, incorporating not the performance but the detailed structural descriptors of the polymers' and MOFs' chemical properties, along with features that capture MOF-polymer interfacial interactions, would further enhance the predictive capability and interpretability of such models. It is important to note that in most studies, the Maxwell permeation model was used, but this classical model has inherent limitations when applied to MMMs, as it assumes ideal spherical fillers with perfect dispersion and interfacial contact within a homogeneous polymer matrix. In real MMMs, effects such as polymer rigidification, interfacial voids, pore blockage, and particle agglomeration significantly deviate from these assumptions, often leading to discrepancies between predicted and experimental permeabilities.

Nearly all high-throughput computational screening studies of MOF/polymer MMMs reported in the literature rely on the rigid MOF assumption during molecular simulations, as it significantly reduces computational cost and time. However, many MOFs are known to exhibit structural flexibility as shown by experiments [39, 40], and incorporating this flexibility into MD

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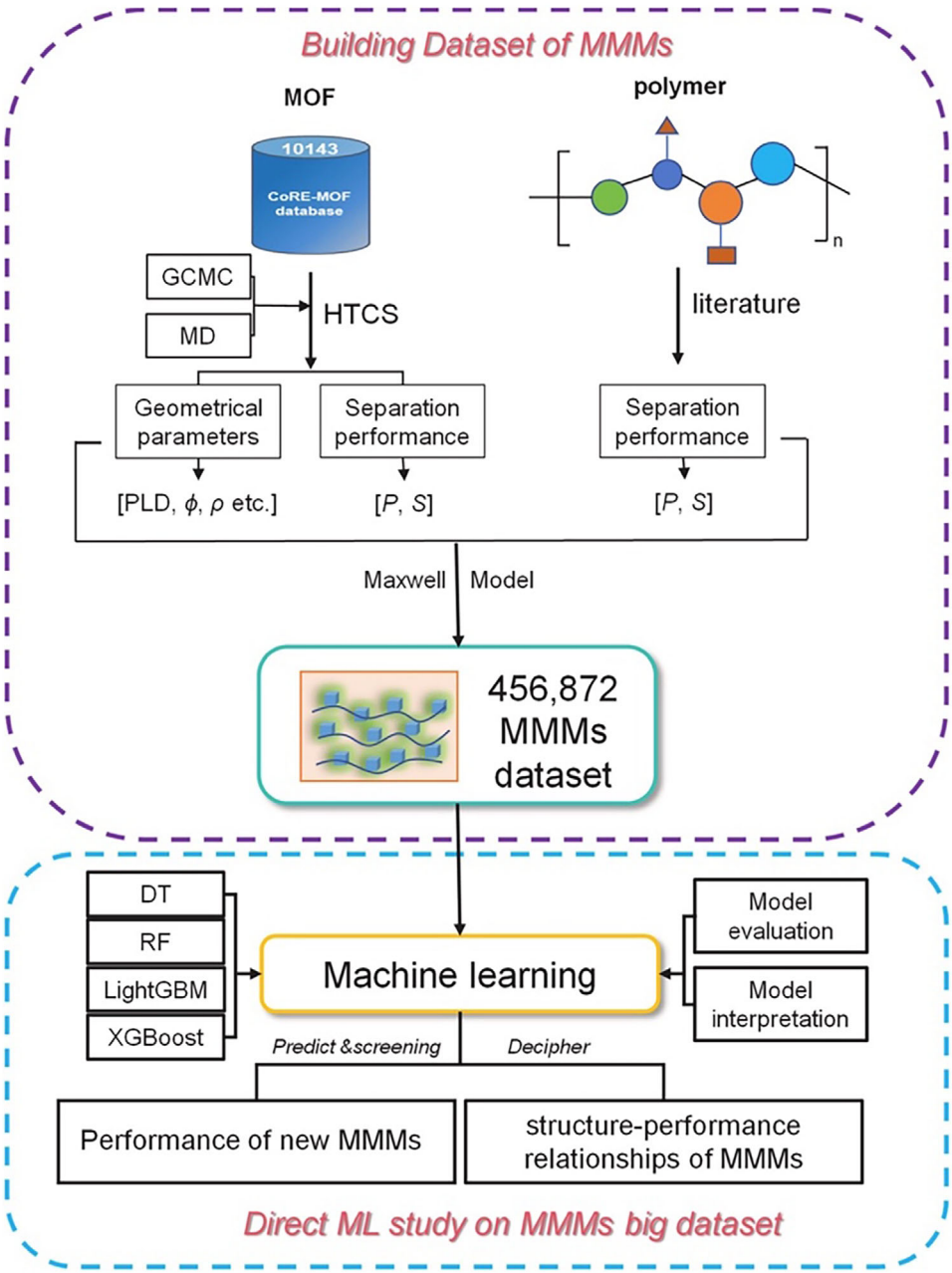


FIGURE 4 | Computational workflow proposed to study large numbers of MOF/polymer MMMs. PLD, ϕ and ρ represent the pore limiting diameter, porosity, and density of the MOF, respectively, whereas P and S are permeability and selectivity. Adapted with permission [38]. Copyright 2025, Elsevier.

simulations yields results that more accurately reflect experimental observations [41]. For instance, several studies [42–44] have reported gas diffusivities that differ by several orders of magnitude when simulations are performed using flexible rather than rigid frameworks. More specifically for MOF membranes, taking framework flexibility into account had little impact on the gas permeability and selectivity of MOFs with large pores, whereas MOFs with narrow pores exhibited more significant changes in gas permeability. To systematically evaluate the influence of framework flexibility on the predicted gas diffusivities in MOFs, a classification ML model was developed by the Sholl group, which reliably predicts whether flexibility significantly impacts molecular diffusion for specific MOF-molecule pairs, including H_2, He, CH_4, N_2, and O_2 [45].

In their follow-up work [46], authors used this classifier ML model together with the Maxwell model for predicting permeability and selectivity of MMMs composed of 6,477 different MOFs and 10 polymers for CO_2/CH_4, O_2/N_2, and He/H_2 separations. As Figure 5 shows, permeabilities predicted from ML and the Maxwell model were in good agreement with the experiments for 98 different MMMs, whereas ML was shown to be less accurate for the MMMs' selectivity predictions. To evaluate the accuracy of ML models used to predict gas diffusivities and loadings in MOFs, molecular simulations were performed on six small-pore MOFs identified by the ML models as promising fillers. Among these, one MOF exhibited gas diffusivities high enough to be reliably measured in simulations. The inability to observe diffusion in the other MOFs raised concerns about the reliability of ML

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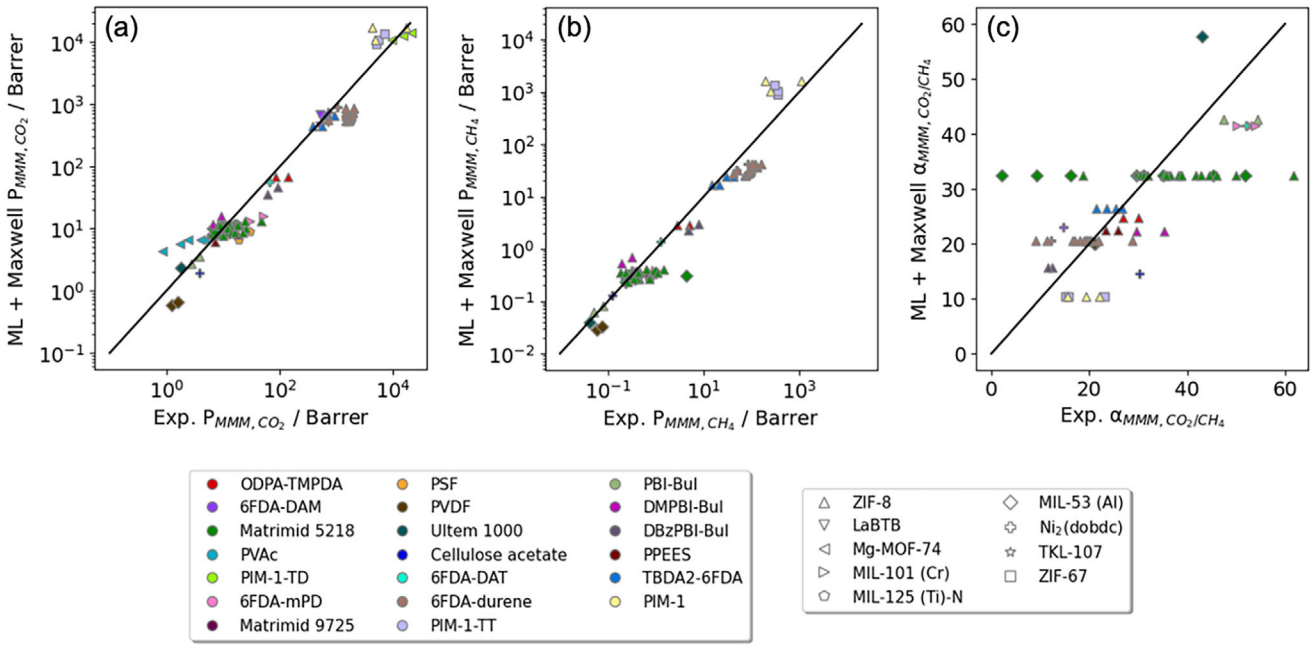


FIGURE 5 | Comparison of ML predictions and experiments for MMMs (a) CO_2 permeabilities, (b) CH_4 permeabilities, (c) CO_2/CH_4 selectivities. Adapted under the terms of the CC BY 4.0 license [46]. Copyright 2025, American Chemical Society.

predictions for small-pore structures. As carefully discussed by the authors, results underscore both the strengths and weaknesses of applying ML in the field of MOF/polymer MMMs. On one hand, ML offers a fast and efficient alternative to classical molecular simulations for studying gas transport properties of MOFs that can be used as fillers in polymers. On the other hand, its effectiveness is constrained by the representativeness of the training data, which may not adequately cover all regions of interest, particularly those involving small-pore MOFs. Thus, future studies focusing on the effect of framework dynamics on the fillers' and hence MMMs' gas separation performance would be very useful.

All studies that we reviewed in this perspective focused on MOF fillers for MMMs. Covalent organic frameworks (COFs), crystalline porous materials built entirely from light elements connected through strong covalent bonds. COFs are not categorized as MOFs, but they have very similar properties to MOFs, such as large pore sizes, high surface areas, and a variety of pore sizes that can be tuned during the synthesis. COFs also have great potential as fillers in MMM applications. For example, Aydin et al., [47] employed molecular simulations to investigate a remarkably large dataset of COF/polymer MMMs, comprising 24,100 COF/polymer MMMs and 4,920 hypoCOF/polymer MMMs. Here, hypoCOFs refer to hypothetical COFs that are computationally generated but have not yet been synthesized. Their results demonstrated that incorporating COF fillers can significantly boost the permeability and selectivity of several polymers, as illustrated in Figure 6. Since COFs have significantly larger pore sizes and porosities than MOFs, many COF/polymer MMMs were predicted to exceed the upper bounds for He/H_2, He/CH_4, He/N_2, H_2/CH_4, H_2/N_2, and N_2/CH_4 separations. Therefore, using the available data of COFs and adapting ML approaches to COF/polymer MMMs will be highly beneficial to unlock the filler performances of COFs for MMM applications.

3 | Conclusion: Future Directions in ML Approaches for MOF/Polymer MMMs

Computational research is set to play an increasingly central role in the design and deployment of MOF/polymer MMMs. By integrating molecular simulations and ML with experimental validation, the field can move beyond trial-and-error approaches toward rational design of high-performance MOF/polymer MMMs. The challenge is not simply to generate more data on MOF/polymer MMMs, but to develop computational frameworks that produce interpretable and realistic predictions for the movement of MMMs seamlessly from the computer to the laboratory scale, and ultimately to industrial use for critical gas separations. Achieving this will require not only advances in integrating ML methods to current experimental and computational approaches but also deeper collaboration between computational scientists and synthetic chemists. Based on these ideas, several challenges, which are indeed some of the future research directions in the field to fully enable novel MOF/polymer MMM design, are discussed as follows:

3.1 | Data for Materials

A fundamental challenge of using ML in the field of membrane science, including the field of MOF/polymer MMMs, is related to the presence and quality of the data. A MMM's performance is directly related to the membrane components' (MOF and polymer) intrinsic properties, such as porosity, fractional free volume, density, surface area, etc [48]. However, this type of information is generally missing or inconsistently reported in the published studies, making the development of ML models difficult. For example, the free volume of the same polymer can be reported differently depending on the synthesis technique, or the pore size of the same MOF can be listed differently in different studies.

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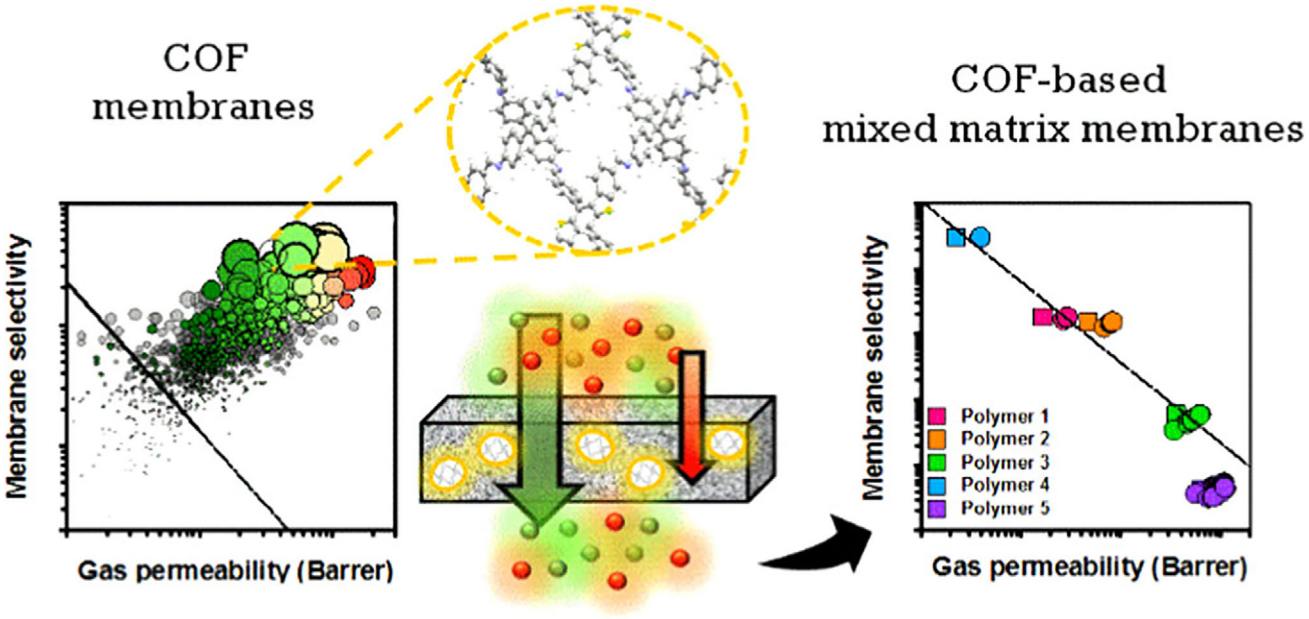


FIGURE 6 | High-throughput computational screening suggests that COFs offer great potential as membranes (on the left) and as fillers in MMMs (on the right) by significantly increasing gas permeability of polymers. The improvement also depends on the type of polymer. Adapted under the terms of the CC BY 4.0 license [47]. Copyright 2022, American Chemical Society.

To ensure reproducibility and comparability across datasets, adopting standardized experimental protocols and clear reporting of measurement conditions is essential. Variations in MMM fabrication, measurement errors, discrepancies in testing equipment, and differences in the operational conditions all contribute to data uncertainty. Incorporating uncertainty information into training data during ML model development, for example, through error-weighted models, can allow predictions to reflect the confidence of underlying measurements. Similar concerns are also valid for the computational studies examining MMMs. Details of the simulation parameters, such as force fields, partial atomic charges, and cycle numbers, should be well-documented so that future users of the ML models can know which type of simulation data was used to train the models and how they can be improved. Overall, to fully harness the potential of data-driven approaches, we need to improve data quality, standardize experimental and computational reporting practices, and expand the availability of comprehensive, high-fidelity datasets.

All the MOF/polymer MMMs studies we reviewed so far have relied on experimentally reported polymer gas permeability data compiled from the literature. Like the case of MOFs, ML offers a promising approach for predicting the gas permeability of polymers when such data are unavailable [49]. For example, artificial neural network regression models were developed to estimate O_2, N_2, CO_2, and CH_4 permeabilities of some glassy and rubbery polymers [50]. Gaussian process regression was applied to predict the gas separation performance of more than 11,000 homopolymers [51], with each polymer represented by a refined molecular fingerprint that encodes the chemical connectivity within its repeating unit. Like the case of hypothetical MOFs and COFs, ML can also be used to identify novel hypothetical polymers. For example, Li and coworkers presented a generalizable ML framework that accurately links polymer chemistry to the gas permeabilities of He, H_2, O_2, N_2, CO_2, and CH_4 [52]. Trained on experimental data, their multitask models not only predict the membranes' performance but also reveal how specific chemical moieties influence permeability and selectivity. After screening over nine million hypothetical polymers, they identified thousands exceeding the upper bounds with CO_2 permeabilities greater than 10^5 Barrer. All these pioneering studies demonstrate that ML can complement experimental efforts by enabling reliable permeability predictions for polymers lacking measured data, thereby expanding the design space for MMMs.

3.2 | Data for Synthesis

Current studies utilized ML only to assess gas permeability and selectivity of the MOF/polymer MMMs. ML can also be used to predict the optimum synthesis conditions of the MMMs. A recent and compelling application of ML involves the development of MOF-derived polymeric membranes for gas separation [53]. MOF-derived polymeric membranes inherit their structural framework from MOFs but cannot be classified as MMMs. In this context, the rational design and precise optimization of the MOF synthesis conditions are critical to ensuring the reproducibility of the MOF precursor material. To address this, an ML-based method introduced previously as Synthesis Condition Finder [54] was adapted to optimize the synthesis parameters of a surface-mounted MOF (SURMOF), incorporating a modified linker to tailor the material's properties for enhanced membrane performance. This approach can be adapted to optimize the synthesis conditions of MOF/polymer MMMs in the future to save a significant amount of time for trial-and-error experiments.

Integrating Bayesian optimization or reinforcement learning with high-throughput experimental data can significantly accelerate

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the optimization of MMM fabrication parameters. Bayesian optimization is ideal for guiding batch or sequential experiments, while reinforcement learning is suited for dynamic, adaptive control of experimental or processing conditions. Both are powerful tools for efficiently navigating complex experimental parameter spaces in MMM fabrication and performance optimization. These methods can efficiently explore multidimensional experimental spaces such as solvent selection, MOF loading, annealing temperature, and casting conditions by learning from previous experimental outcomes to identify combinations that minimize interfacial defects and maximize separation performance.

Capturing the complex nature of MOF-polymer interfaces is crucial for realistic MMM performance predictions. ML can offer a promising framework to address this challenge by learning structure-property relationships from both simulations and experiments that reflect interfacial phenomena. For example, relevant features such as polymer rigidification, non-selective voids, pore blockage, and functional-group compatibility can be represented through descriptors derived from MD simulations (e.g., local density profiles, accessible pore volume changes, interfacial binding energies) and experimental data (by using Atomic Force Microscopy, Fourier Transform Infrared Spectroscopy). ML models trained on such descriptors, especially graph-based or physics-informed approaches, can establish correlations between interfacial structure and gas transport performance, thus enabling quantitative prediction and rational control of interfacial defects. Development of ML-driven surrogate models to rapidly predict interfacial properties for large libraries of filler-polymer pairs will be important to guide the experimental synthesis efforts.

3.3 | Novel Designs

Predicting durability-related properties of MMMs, such as plasticization resistance, physical aging, and hydrothermal stability, is essential for the practical deployment of these hybrid membranes in industrial applications. While the current literature solely focuses on predicting gas separation performance of MMMs, similar ML frameworks can indeed be extended to these metrics. For instance, ML models trained on features collected from the literature such as polymer type and chemistry, experimentally observed plasticization pressures can estimate the pressure at which plasticization is likely to occur for an MMM. Incorporating such durability-related targets into future ML models will also be crucial for developing MMMs that combine high selectivity with long-term operational stability.

While still in early stages, generative ML models can create MOF-polymer combinations optimized for target separations [55]. These models can integrate structural generation with performance prediction, aiming to propose not only the MOF but also the compatible polymer and loading fraction. Current ML studies have primarily focused on predicting single-component gas permeability through MMMs. However, in industrial applications, gases are typically present as mixtures, so models capable of predicting the separation performance of MMMs for various mixture compositions would be highly valuable. Similarly, investigating membrane performance at pressures and temperatures beyond conventional conditions (1 bar, 298 K) can provide insights into the optimal operating conditions for achieving high gas separation efficiency. Beyond separation performance, application-driven success metrics shaping the future of MMMs include scalability, long-term stability, and cost. These aspects, such as optimizing filler loading to balance stability and economic feasibility, can be systematically studied by using ML.

It is worth noting that current ML studies have focused on MOFs rather than COFs, largely because the number of available MOFs, both experimental and hypothetical, is considerably greater, which is advantageous for ML models that benefit from the large datasets. The superior chemical robustness of COFs compared to MOFs may be another factor motivating the use of ML-guided filler screening toward COFs. Currently, we are aware of about 70,000 hypothetical COFs and millions of hypothetical MOFs. When working with these hypothetical materials, the synthesizability of the proposed structures must also be considered. Some of the most promising fillers that are expected to yield high-performance MMMs may be challenging to synthesize, or the synthesized versions may have stability issues. Therefore, integrating ML tools to evaluate the synthesizability and stability of hypothetical fillers for the design of novel MMMs represents an important research direction.

3.4 | Bridging Scales

A persistent gap exists between molecular-level insights obtained from molecular simulations and experimentally measurable properties. Validating predictions obtained from ML requires close collaboration with experimentalists, along with iterative workflows where simulations guide experiments and experimental results refine the ML models. Community-driven efforts to create open membrane databases will be essential for accelerating ML-driven MMM research. Given that robotic synthesis and automated testing are already being applied to the production of crystalline materials [56, 57], these approaches could be adapted for MOF/polymer MMM fabrication. This would enable experimental validation of computational predictions and refinement of models, potentially accelerating the discovery-to-deployment pipeline.

Overall, ML holds great potential for reducing the experimental and computational costs associated with studying MOF/polymer MMMs. However, it is equally important to recognize that the unnecessary or improper use of ML can be highly problematic. Applying ML with little or no validation, particularly in cases where, for example, classical computational methods are already well-established and may even perform better, poses a significant risk. Molecular simulations have long been used to investigate polymer and MOF membranes, and experts in this field generally know what to expect and how to interpret the outcomes based on their experience. With ML, however, this is not always the case, as interpretable applications of ML in this domain are still in their early stages. Therefore, careful integration of ML with the state-of-the-art experimental and computational approaches will hold great promise for accelerating the design, discovery, optimization, and deployment of MOF/polymer MMMs, paving the way toward more efficient and targeted gas separation technologies.

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Conflicts of Interest

The authors declare no conflict of interest.

Data Availability Statement

Since this is a Perspective paper, no new data is generated for this manuscript.

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