Machine Learning-Assisted Design and Chemical Screening of Metal–Organic Frameworks for Enhanced Carbon Dioxide Capture and Electrochemical Energy Storage Applications

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Abstract

Because of the rising concentration of CO_2 in the atmosphere and the increasing demand for energy technologies that can improve energy storage efficiency, there has been a renewed interest in developing advanced functional materials that can help address some of the world's energy and environmental challenges. Metal-organic frameworks (MOFs), as a class of newly developed crystalline porous coordination polymers (made up of metal ions/clusters covalently bonded together using organic ligands), are a promising candidate to potentially provide solutions to these challenges due to their high specific surface area, tunable pore size, chemical diversity, and structural diversity. Coordination chemistry, metal-node characteristics, and linker functionalities (i.e., coordination chemistry and qualities of the metal nodes and linker groups) will play a large role in influencing the MOFs' adsorptive ability, electrochemical performance, and stability. Traditional approaches to finding and optimizing new MOFs require large amounts of time, money, and labor. To streamline this process, a computational chemistry framework was developed to identify and assess new high performance MOFs for application to CO2 capture and electrochemical energy storage. The framework utilized a database containing 2,500 unique MOF structures curated from well-established databases and published literature. The structural descriptors of these MOFs (physical, chemical, and mechanical property values, such as surface area, pore volume, framework density, metal electronegativity, metal coordination, thermal stability, electrical conductivity, and ion diffusion) were used to aid model training. Machine learning models were created using common algorithms (Random Forest, Support Vector Machine, Gradient Boosting, and Artificial Neural Networks) to predict the maximum amount of CO2 that can be adsorbed and/or the maximum specific capacitance of a MOF per gram of MOF material. Random Forest was the best algorithm, with an R2 value >0.90 for both target values. The top five important determining factors in predicting the MOFs' adsorptive and capacitive properties were shown to be surface area, pore volume, electronegativity of the metal nodes, functionality of the linkers, and stability of the framework. A chemical explanation of MOF adsorptive and electrochemical properties was determined using Lewis acid-base interactions

between the open metal sites of the MOFs and CO2 molecules, along with linker functionalization (i.e. adding specific functional groups to the organic linkers). The addition of redox-active transition metal ions and conjugated organic linkers allowed for increasing charge storage capacity and use of electroactive materials for conducting electrons. Through the use of AI-based virtual screening methods, it was possible to identify hypothetical MOFs with predicted CO2 capture capacities >9 mmol/g and specific capacitances approaching 500 F/g. The successful combination of coordination chemistry, computational chemistry techniques, and AI is shown to be an efficient means of in vitro design of next generation MOFs. This work will be utilized to develop advanced porous materials for sustainable technologies that capture CO2 and produce high-performance energy storage devices.

Keywords: Metal–Organic Frameworks, Coordination Chemistry, Carbon Dioxide Adsorption, Energy Storage, Computational Chemistry, Artificial Intelligence, Machine Learning, Materials Informatics.

1. Introduction

One of the two major challenges that science and technology face during the twenty-first century is the need to manage increasing levels of carbon dioxide (CO_2) in our atmosphere and the need for sustainable forms of energy worldwide. Fossil fuels played an extremely important role in this rise of carbon dioxide levels through the development and use of fossil fuel-based systems, rapid industrialization, and the rapid growth of cities and the urbanization of Earth's population. The rise of greenhouse gas emissions through these processes has led to such undisputed changes that global climate change, ocean acidification, biodiversity losses, and long-term ecosystem degradation will occur in the foreseeable future. Climate Assessment reports produced recently reveal that atmospheric concentrations of CO_2 are over 420 parts per million (ppm), with concentrations at 420 ppm representing the highest recorded level of carbon dioxide in the atmosphere in history. This large increase of greenhouse gases in our atmosphere is clearly linked to major increases in global temperatures, major increases in extreme weather events, major declines in worldwide biodiversity, and long-term degradation of Earth's environment (Li *et al*., 1999, Yaghi *et al*., 2003, Furukawa *et al*., 2013). As a result, developing advanced materials and processes for carbon capture/sequestration and developing advanced technologies for the efficient and safe storage of energy have been designated by major government/military organizations and as such, has become an area of priority research globally. In this context, MOFs (Metal-Organic Frameworks) are one type of class of advanced functional materials of many types of advanced functional material types that has received significant scientific interest due to their unique structural and chemical properties. MOFs are three-dimensional crystalline porous materials met from the self-assembly of metal ions (or metal atomic clusters), and multifunctional organic ligands. Combination of metal ions (or metal atomic clusters) and multifunctional organic ligands form extremely large, highly ordered three-dimensional structural frameworks with extreme amounts of surface area (≥10 m^2/g)(radical structural diversity) creating the potential for tunable holes in the structural frameworks or pores, as well as controllable functions (radical chemical functionality). Since the pioneering work of Yaghi and co-workers in the late 1990s on porous coordination materials (2003), tens of thousands of unique MOF structures have been developed using 200+ different metal ionic species/atomic clusters, and thousands of different organic ligands for gaseous storage and separation, catalysis and chemical sensing, drug delivery, environmental remediation, and energy storage. (Li *et al*., 1999; Yaghi *et al*., 2003; Furukawa *et al*., 2013).Coordination chemistry is the foundation for how Metal Organic Frameworks (MOF) are formed and developed. Metal ions act as the Lewis acids and organic ligands act as Lewis bases that donate pairs of electrons to create what is called a coordinate covalent bond. The formation of a vast range of different framework topologies depends on coordination geometry; oxidation state; electronic configuration; and ligand denticity. Commonly used metals in developing MOFs are: Zn^{2+};

Cu^{2+}; Co^{2+}; Ni^{2+}; Fe^{3+}; Cr^{3+}; Al^{3+}; Zr^{4+}. Commonly used organic linkers are: terephthalic acid; trimesic acid; imidazole derivatives and pyridine based ligands or multifunctional aromatic compounds. The assortment of both metal nodes and organic linkers used allows for a tremendous amount of flexibility when tailoring different framework properties to suit specific needs of users or to perform to specific industrial applications. One attractive aspect of MOFs is the very high specific surface area of most MOFs. Surface areas of many MOFs are greater than 7000 $m^2 g^{-1}$ which are much greater than all traditional porous materials including activated carbon, dielectric silica gels and zeolite (Furukawa *et al*., 2010). MOF exhibit exceptionally high surface areas in combination with tunable pore sizes that provide efficient adsorption, storage and transport of gases and ions. Therefore, MOFs have become highly qualified candidates for CO_2 capture, hydrogen fuel cell storage, methane fuel cell storage and electrochemical energy storage applications.

From a chemistry standpoint, the outstanding performance that MOFs have for capturing CO_2 is dictated not only by their porosity and density, but also by their ability to form host–guest interactions with CO_2. CO_2 is a linear molecule with a very high quadrupole moment and is a weak Lewis base.The relation between electrostatic attraction among CO2 molecules and the unfilled coordination sites of metal ions within the framework of Metal-Organic Frameworks (MOFs) is an important factor that influences how CO2 is adsorbed onto MOFs. Lewis acidic sites at the metal ions that are directly incorporated within the MOF Framework will have a strong electron attraction to the oxygen atoms in the CO2 molecule. The strength of the electrostatic attractions between CO2 molecules and the metal coordination sites will lead to a greater affinity to adsorb onto and capture CO2 molecules.

The unfilled metal coordination sites in the MOFs in addition to the chemical functionalization of the organic linkers in the framework also have a significant impact on how CO2 is adsorbed. Amino (-NH2), Hydroxyl (-OH), Carboxyl (-COOH) and Pyridyl functional groups can be added to the MOF either during the synthesis process or after the fact (post-synthesis). The presence of functional groups provides local electric fields and hydrogen bonding interactions with MOFs which increases the affinity for CO2 adsorption. A number of articles indicate that MOFs with amine functional groups have significantly greater CO2 adsorption properties than MOFs that do not contain functional groups due to the cooperative mechanisms by which the CO2 are adsorbed on the MOF, as well as the acid/base interactions put in place by the functional group (Sumida *et al*., 2012).

The performance longevity of a MOF is also an important consideration in how a MOF can be chemically utilized. Carbon capture processes found in many manufacturing facilities usually are subject to the MOF being at elevated temperatures, under conditions of high water vapor, and being exposed to other high chemically aggressive materials. Therefore, in order to ensure that a MOF structure maintains its performance at the necessary level, the MOF framework must also be a highly stable structure. MOFs can be classed as stable or unstable structures by utilizing the Pearson Hard and Soft Acid Base (HSAB) Theory. MOF structures with strong coordination bonds created by the hard metals (i.e. Zr4+, Al3+, and Fe3+) and the hard oxygen donating ligands result in highly stable coordination bonds that are resistant to hydrolysis and thermal degradation (i.e. UiO-66, MIL-101, and other related Zr-based MOF frameworks) and provide a great level of stability for chemical and thermal conditions related to industrial processes.While softer metal / ligand interactions may provide improved flexibility and porosity, they frequently possess lower hydrolytic stability.

MOFs have also gained considerable attention for use in electrochemical energy storage technologies beyond their environmental application. As such, the growing utilization of renewable energy sources (solar and wind) creates a need for energy storage systems that can provide ready access to stored energy for addressing the variable nature of energy production from these sources. Thus, advanced electrode materials with large surface area, good conductivity, fast ion transport, and long-

term cycling stability will be required by batteries, supercapacitors, and hybrid energy storage devices. The ability to tune the chemistry and porous structure of MOFs makes them a very desirable choice for using in these applications.

The electrochemical characteristics of MOFs are controlled by the two charge storage mechanisms described above: Electrical Double Layer Capacitance (EDLC), in which electrolyte ions are stored electrostatically at the internal surface of the porous (framework) of a MOF; and Pseudocapacitance, which results from the reversible Faradaic oxidation-reduction reactions that occur at the redox-active metal centers of a MOF. The transition metals (Co, Ni, Mn, and Fe) present in the MOFs can change their oxidation state reversibly, thereby contributing to their capacity for charge storage and energy density.

In addition to this, recent work illustrating the outstanding electrochemical activity of MOF-derived materials in energy storage applications is due in large part to the hierarchical pore structure, the presence of high-active surface areas, and abundant redox-active sites, as well as having conductive organic linkers containing extended π-conjugated systems that enhance electron transport through the framework of the MOFs. The strong π–d orbital interactions that occur between organic ligands and the transition metal centers in the framework of the MOFs also contribute to the enhanced conductivity and electrocatalytic activity of these materials.A rational approach to designing MOF structures via the chemical modifcation of the MOFs will be successful for creating next--generation electrode materials for super capacitors and rechargeable batteries.

Expanding the capabilities of computational chemistry to produce faster and less expensive searches for and optimizations of Metal-Organic Frameworks (MOFs) through computational modelling techniques, e.g., DFT, MD, Monte Carlo algorithm, GCMC Calculation, etc. Traditional methods of creating and analyzing MOFs are typically labor-intensive, expensive, and time-consuming. Thus, it may take many months to years of work for researchers to create/design, produce, and analyze a single MOF structure. Due to this, researchers are using Computational Modelling Methods to predict the properties of MOFs prior to actually testing the MOFs experimentally. Using DFT to examine the properties of MOFs at the molecular level is perhaps the most common methodology used in the study of MOF properties. Via DFT calculations, researchers can produce data regarding the electronic structure of a MOF; i.e., charge distribution, adsorption energy, the way that molecular orbitals interact, and the nature of the bond (i.e., covalent, ionic, lossy, etc.) between the metal and the ligand. DFT analysis also enables researchers to determine where on the framework maximum amounts of carbon dioxide will be absorbed and how strong the intermolecular force is acting on the carbon dioxide and the surface(s) of the MOF's framework(s). Additionally, the adsorption energy produced by DFT modelling of the carbon dioxide will provide researchers with a measure of the adsorption efficiencies and selectivity (i.e., how the carbon dioxide will differentiate from other adsorbate molecules) of the MOF's framework(s). Open metal sites and functionalized ligands can increase the adsorption of carbon dioxide through increased electrostatic interaction between carbon dioxide and the functionalized ligand and via electron transfer processes between carbon dioxide and the open metal site of the MOF (Boyd *et al*., 2019; Coudert, 2015).The Grand Canonical Monte Carlo (GCMC) method has emerged as another important computational tool used in evaluating gas adsorption upon porous materials. GCMC simulations allow for the prediction of the adsorption isotherm, capacity, selectivity and diffusion under a variety of temperature and pressure conditions. These simulations are particularly beneficial for screening the many libraries of hypothetical metal organic frameworks (MOFs) prior to experimental synthesis. Previous studies have shown a close correlation between the GCMC predicted adsorption capacities and those measured experimentally, demonstrating that computational screening approaches can be trusted in discovering new materials (Boyd *et al*., 2019; Jablonka *et al*., 2021).

With the advent of materials informatics, the field of materials science has changed once again. Materials informatics utilizes computational chemistry, materials databases, statistics, and artificial intelligence to accelerate the discovery of novel materials. Instead of relying on an experimental and trial-and-error basis alone, researchers can utilize large datasets to discover hidden relationships between structural descriptors and materials properties. The existence of several large-scale databases (Cambridge Structural Database (CSD), CoRE MOF Database, Materials Project, Open Quantum Materials Database (OQMD)) has opened numerous new doors to discovering new materials via data-driven methods (Chung *et al*., 2014; Jain *et al*., 2013).

AI and ML have become two more new essentials in computational methods that are capable of discovering nonlinear relationships in very large datasets. ML algorithms have the capability to examine thousands of material structures simultaneously using the physicochemical descriptors of the materials to create predictive performance based on these structures. Unlike traditional statistical methods, ML methods are able to identify multidimensional relationships between the variables, which allow them to produce very accurate predictive models.As a result, artificial intelligence (AI) has become more essential to computational chemistry, catalysis, drug development, materials science and environmental engineering than ever before (Butler *et al*., 2018; Agrawal and Choudhary, 2019). Machine learning algorithms performed well in predicting the adsorption characteristics of metal organic frameworks (MOFs), their thermal stability, mechanical strength, catalytic activity and electrochemical performance. Many different types of algorithms such as random forests (RFs), support vector machines (SVMs), gradient boosting (GB), extreme gradient boosting (XGBoost), artificial neural networks (ANNs) and deep learning architectures have all produced good prediction performance with MOF data sets. These ways to use machine learning can greatly reduce the number of laboratory experiments and computational simulations required to identify new MOF candidate materials on which to focus research efforts (Singh *et al*. 2008, Moosavi *et al*., 2020; Schmidt *et al*., 2019). Random forest, in particular, has gained popularity as a machine learning approach due to its robustness, interpretability, and ability to model non-linear relationships. Random forests work by taking an ensemble of decision trees that each predict a value for an observation, use that collection of predictions as input to develop a statistical model for the prediction of future values for the same observation, and combine those predictions together to improve the predictions of the individual trees (Breiman, 2001; Moosavi *et al*., 2020) While ANN technology has recently attracted considerable research attention for its ability to learn complicated and non-linear structure-property relationships directly from the data.The performance of predicting adsorption capacities and electrochemical properties of porous materials has been remarkable with deep neural networks (DNNs) with multiple hidden layers (Badruddin and Singh. 2009, LeCun *et al*., 2015; Rosen *et al*., 2021). The successful application of Machine Learning (ML) in searching for new materials greatly relies upon both the accurate identification and selection of appropriate descriptors. Descriptors can be defined as a numerical quantity that measure and portray the structure, chemistry, electronic nature and physical/chemical characteristics of a material. The descriptors associated with Metal-Organic Frameworks (MOFs) can generally be divided into two categorically different types, structural and chemical. Structural descriptors include surface area, pore volume, pore diameter, density, void fraction, and crystal symmetry. The mentioned structural parameters define the characteristics that relate to how accessible and how well a substance can move through the pore system and/or be adsorbed into porous materials. Structural descriptors alone do not fully describe or account for the complex nature of adsorption behavior and electrochemical activity of MOFs.

Chemical descriptors create additional knowledge of the fundamental chemistry that affects properties of materials. Metal electronegativity, oxidation state, atomic radius, coordination number, electronic configuration and metal-node identity are descriptors of materials that best describe how

such materials will adsorb a substance or exhibit electrochemical behavior. For example, metals having greater charge density will typically exhibit stronger electrostatic interactions to carbon dioxide molecules, resulting in greater adsorption enthalpy and greater adsorption; thus, producing higher quantities of carbon dioxide adsorbed. In a similar manner, metal/transition metal centers that demonstrate reversible oxidation/reduction behavior can greatly enhance the pseudocapacitive performance of a material (Simon & Gogotsi, 2020, Rafi *et al*., 2025).

Organic linker chemistry also has a large impact on the overall performance of the framework. The ability of the framework to polarize, conduct electricity and exhibit selective adsorption is significantly influenced by the length of the conjugation of the aromatic rings, whether the aromatic rings are connected to each other through oxygen-containing heteroatoms or through other means, as well as the electronic properties of the functional groups that connect the aromatic rings to the MOF frame. Groups that donate electrons like those with amine groups make Metal-Organic Frameworks (MOF) better at attracting acid gases like CO_2. At the same time, some ligands that have sulphur atoms help make MOFs better at conducting electricity and making chemical reactions. Examples of this come from research by Li and others in 2016 where they used the Hammett constant to show that the electronic effects of different kinds of linkers can relate to how they hold onto gases or their ability to catalyse chemical reactions.

In the past couple of years, other researchers have found materials scientists can use information from Hard and Soft Acid-Base (HSAB) theory as a tool to predict the level of stability of a framework. By combining hard metal ions like Zr or Al, which normally create stable frameworks, with ligands that donate oxygen, researchers can generally create very stable frameworks. Whereas in cases of softer metal ion to ligand interactions, the resulting framework is generally more flexible and has greater porosity. These kinds of chemical descriptors are an important part of the mechanical understanding of how materials work in conjunction with their physical and structural parameters, as well as enhancing performance in machine learning-type processes (Pearson, 1963; Furukawa *et al*. 2013 Ahmad *et al*., 2022).

Although there have been tremendous advancements in the use of machine learning tools to aid materials development, there are many challenges to face. Many of the materials frameworks used thus far use only structural parameters and do not include adequate detail on the coordination of the metals involved, how the electronic structures of the metals or ligands contribute to the stability of that material, or how the overall functionality of the linker will affect gas retention or energy retention. Therefore, many of the models created using machine learning are "black boxes" in that they can give accurate predictions, but do not explain how the prediction was made chemically. Bridging the gap between AI and Coordination Chemistry is an essential step toward the development of interpretable, utilitarian predictive models.

Our study integrates AI, computational chemistry, coordination chemistry and materials informatics in order to create predictive models of metal organic frameworks (MOFs) that can be used to find new materials for CO_2 capture and electrochemical energy storage. By integrating structural and chemical descriptors into the machine learning model, the goal of this work is to determine meaningful structure-property relationships that can be used to rationally design future generations of porous materials. The goals of our work are as follows: (i) to develop machine learning models for predicting carbon dioxide capacities and specific capacitances; (ii) to analyse structural-charateristics vs functional chacteristics to evaluate the importance of chemical descriptors compared to structural descriptors; (iii) to derive a better understanding of the processes whereby chemical adsorption occurs and how they relate to how energy can be stored; and (iv) to provide a basis for identifying

new MOFs that have been characterised as showing superior capabilities to use machine aided means of visual screening.

2. Materials and Methods

2.1 Study Design

This study used a computational chemistry and artificial intelligence framework to create a way to find and screen Metal Organic Frameworks (MOFs) that could be used to capture carbon dioxide and store electrical energy. This approach used the principles of coordination chemistry with the fields of materials informatics, machine learning algorithms, and computational screening in order to create structure/property relationships to describe how well MOFs would absorb carbon dioxide (CO_2) and also provide electrochemical energy storage performance.

The overall methodology consisted of six steps:

1. assemble a dataset

2. extract descriptors from the dataset

3. characterize the chemical structure and properties of each dataset member

4. develop machine learning models that relate the extracted descriptors to CO_2 absorption and electrical energy storage performance

5. determine which descriptors were the most important in determining CO_2 absorption and electrical energy storage performance, and

6. conduct a computer-assisted virtual screen of possible hypothetical MOF designs. The final framework was constructed to help determine what chemical and structural factors influence CO_2 absorption and energy storage performance the most.

2.2 Collection of MOF Dataset

An extensive compilation of obtained experimental and computed MOFs was created using respected databases of materials around the world and the relevant peer-reviewed published resources. Information on the structure, physical and chemical properties, adsorption and electrochemical data were compiled from different sources:

- CoRE MOF Database

- Cambridge Structural Database (CSD)

- Materials Project Database

- Open Quantum Materials Database (OQMD)

- Published literature on carbon capture and energy storage applications

Initially, approximately 5,000 MOF structures were collected. Duplicate records, incomplete entries, and inconsistent datasets were removed during preprocessing. After data cleaning and validation, a final dataset comprising 2,500 representative MOFs was selected for machine learning analysis.

Table 2.1 Dataset Summary

Parameter	Value
Total MOFs collected	5,000
Final dataset	2,500
Structural descriptors	15
Chemical descriptors	10
Carbon capture records	2,500
Energy storage records	2,500

2.3 Chemical Basis of Descriptor Selection

Unlike conventional machine learning studies that rely solely on geometric descriptors, the present investigation incorporated both structural and chemical descriptors to improve mechanistic interpretation.

2.3.1 Structural Descriptors

The structural descriptors selected included:

- Surface Area ($m^2\ g^{-1}$)
- Pore Volume ($cm^3\ g^{-1}$)
- Pore Diameter (nm)
- Density ($g\ cm^{-3}$)
- Void Fraction (%)
- Crystal Symmetry Index
- Ion Diffusion Rate

These descriptors directly influence adsorption capacity, gas diffusion behavior, electrolyte accessibility, and electrochemical performance.

2.3.2 Chemical Descriptors

The chemical descriptors were selected based on coordination chemistry and adsorption chemistry principles.

The descriptors included:

- Metal Atomic Radius

- Metal Electronegativity
- Oxidation State
- Coordination Number
- Metal Node Identity
- Open Metal Site Density
- Linker Aromaticity Index
- Linker Functional Groups
- Heteroatom Content
- Framework Stability Index

These parameters were expected to influence adsorption affinity, charge transfer processes, framework stability, and redox activity.

Table 2.2 Chemical Descriptors Used in Machine Learning Models

Descriptor	**Chemical Significance**
Metal Electronegativity	CO_2 polarization ability
Oxidation State	Redox activity
Coordination Number	Framework geometry
Open Metal Sites	Lewis acidity
Linker Aromaticity	Electron transport
Nitrogen Content	CO_2 affinity
Sulfur Content	Conductivity enhancement
Framework Stability Index	Chemical durability
Metal Radius	Pore architecture
HSAB Compatibility Score	Stability prediction

2.4 Coordination Chemistry Analysis

The coordination chemistry of each framework was evaluated to understand metal–ligand interactions and framework stability.

Metal ions were classified according to:

- Electronic configuration
- Oxidation state
- Coordination geometry
- Hard–Soft Acid–Base (HSAB) character

According to Pearson's HSAB principle, hard metal ions such as Zr^{4+}, Al^{3+}, and Fe^{3+} form highly stable coordination bonds with oxygen donor ligands, whereas softer metals such as Co^{2+}, Ni^{2+}, and Zn^{2+} exhibit stronger interactions with nitrogen-containing ligands.

Framework stability was assessed through metal–ligand bond strength and reported thermal decomposition temperatures.

2.5 Computational Chemistry Analysis

2.5.1 Density Functional Theory (DFT)

Quantum chemical calculations were considered for evaluating adsorption behavior and electronic properties of representative MOFs.

DFT analysis was used to investigate:

- Adsorption energy
- Charge distribution
- Electronic density
- Metal–ligand bonding
- Frontier molecular orbitals

The generalized gradient approximation (GGA) utilizing the Perdew–Burke–Ernzerhof (PBE) functional was selected because of its suitability for porous materials and adsorption studies

2.5.2 Grand Canonical Monte Carlo (GCMC) Simulations

Carbon dioxide adsorption behavior was analyzed through Grand Canonical Monte Carlo simulations.

Simulation conditions:

Temperature = 298 K

Pressure Range = 0–10 bar

Adsorbate = Carbon Dioxide (CO_2)

The simulations estimated:

- Adsorption isotherms
- Adsorption selectivity
- Gas loading capacity

• Heat of adsorption

The calculated adsorption capacities served as reference values for machine learning model development.

2.6 Adsorption Chemistry Evaluation

The adsorption behavior of CO_2 was interpreted using Lewis acid–base interactions.

Open metal sites within MOFs act as Lewis acidic centers capable of interacting with the quadrupole moment of carbon dioxide molecules.

The following adsorption mechanisms were considered:

Physical Adsorption

• Van der Waals interactions

• Electrostatic interactions

• Pore confinement effects

Chemical Adsorption

• Lewis acid–base interactions

• Hydrogen bonding

• Coordination interactions

• Charge transfer effects

Particular emphasis was placed on amino-functionalized frameworks because amine groups are known to enhance CO_2 capture through acid–base chemistry.

2.7 Electrochemical Characterization Parameters

The electrochemical suitability of MOFs for energy storage applications was evaluated using descriptors associated with charge transport and redox activity.

Parameters included:

• Electrical Conductivity

• Specific Capacitance

• Charge Transfer Resistance

- Ion Diffusion Rate
- Redox Activity

The energy storage mechanisms were classified into:

Electrical Double Layer Capacitance (EDLC)

Electrostatic accumulation of electrolyte ions at the electrode–electrolyte interface.

Pseudocapacitance

Reversible Faradaic oxidation–reduction reactions involving transition metal centers such as:

Co^{2+}/Co^{3+}

Ni^{2+}/Ni^{3+}

Fe^{2+}/Fe^{3+}

Mn^{2+}/Mn^{3+}

These redox-active species contribute significantly to charge storage capacity.

2.8 Data Preprocessing

Prior to model development, all datasets underwent preprocessing.

Missing Value Treatment

Missing values were replaced using median imputation.

Outlier Removal

Outliers were detected using the Interquartile Range (IQR) method.

Feature Normalization

Numerical variables were normalized using Min–Max scaling.

Data Partitioning

Training Dataset = 80%

Testing Dataset = 20%

This approach ensured unbiased model evaluation and reduced overfitting.

2.9 Machine Learning Algorithms

Four machine learning algorithms were employed.

Random Forest (RF)

Advantages:

• High predictive accuracy

• Nonlinear relationship modeling

• Feature importance interpretation

Support Vector Machine (SVM)

Advantages:

• Effective for high-dimensional datasets

• Strong generalization capability

Gradient Boosting (GB)

Advantages:

• Sequential error minimization

• Reduced bias

Artificial Neural Network (ANN)

Network Architecture:

Input Layer = 25 Neurons

Hidden Layer 1 = 64 Neurons

Hidden Layer 2 = 32 Neurons

Output Layer = 1 Neuron

Activation Function = ReLU

Optimizer = Adam

Learning Rate = 0.001

Epochs = 200

Batch Size = 32

2.10 Target Variables

Two principal target variables were selected.

Carbon Capture Performance

Measured as:

CO_2 Adsorption Capacity (mmol g^{-1})

Energy Storage Performance

Measured as:

Specific Capacitance (F g^{-1})

These indicators are widely accepted for evaluating MOF suitability for carbon capture and electrochemical applications.

2.11 Feature Importance and Explainable AI

To improve chemical interpretability, feature importance analysis was performed using:

- Random Forest Feature Ranking
- SHAP (SHapley Additive Explanations)

The analysis identified descriptors contributing significantly to:

- CO_2 adsorption
- Framework stability
- Charge transport
- Electrochemical performance

This approach helped bridge the gap between machine learning predictions and chemical understanding.

2.12 AI-Assisted Virtual Screening of Novel MOFs

Following model validation, virtual screening was conducted on a computational library of hypothetical MOFs.

Predicted properties included:

- CO_2 Uptake Capacity
- Specific Capacitance
- Framework Stability
- Conductivity
- Adsorption Selectivity

Only the highest quality candidates were chosen for chemical processing.

Those who will produce the most significant results for carbon capture and energy storage applications in terms of capabilities were the ones we focused on most heavily in selecting candidates (i.e., the use of amine functionalized frameworks based on zirconium, as well as cobalt/nickel-based redox active frameworks).

2.13 Statistical Analysis

All analyses were performed using Python programming language.

Software and libraries used:

- Scikit-Learn
- TensorFlow
- Pandas
- NumPy
- Matplotlib

Model validation was performed using five-fold cross-validation.

Statistical significance was accepted at:

$p < 0.05$

The robustness, reproducibility, and predictive capability of the developed models were evaluated using R^2, RMSE, and MAE metrics.

3. Results

3.1 Characteristics of the MOF Dataset

The study used a data set of 2500 Metal-Organic Frameworks collected from literature and databases. These frameworks had diversity in topology, inorganic and organic composition, and function. The frameworks included zirconium, copper, cobalt, nickel, iron, and zinc. The organic linkers also had a wide range of functional subcategories. Surface area for the frameworks varied between 850-7200

m^2/g. Pore volume for each framework varied between $0.35cm^3/g$ to $2.75cm^3/g$. As for the electronegativity for the metallic nodes; this value varied depending on the node as well. Indices of framework stability also varied; this reflects the strength of the bonds with the metals (nodes) and the type of coordination chemistry.

Table 3.1 Descriptive Statistics of Major Structural and Chemical Variables

Variable	Minimum	Maximum	Mean	SD
Surface Area ($m^2\ g^{-1}$)	850	7200	3154	1120
Pore Volume ($cm^3\ g^{-1}$)	0.35	2.75	1.42	0.53
Density ($g\ cm^{-3}$)	0.21	1.25	0.62	0.18
Metal Electronegativity	1.22	2.28	1.71	0.24
CO_2 Uptake ($mmol\ g^{-1}$)	2.1	10.2	6.8	1.7
Specific Capacitance ($F\ g^{-1}$)	110	520	325	85

The broad distribution of physicochemical and chemical parameters provided a robust dataset for machine learning model development and feature interpretation.

3.2 Coordination Chemistry and Framework Stability

The analysis of metal-ligand interactions showed that the framework stability was highly influenced by the characteristics of metal nodes and organic ligands. The frameworks that were made using zirconium had the highest average stability scores because of the presence of strong Zr-O coordination bonds. The frameworks that contained iron and aluminium also had very good thermal stability due to the favourable hard acid-hard base interactions as dictated by Pearson's HSAB theory.

On the other hand, the frameworks that utilized zinc and cobalt exhibited a low degree of rigidity, which provided them with larger pore volumes but a slightly reduced degree of hydrolytic stability. Frameworks utilizing nitrogen-rich imidazolate ligands were better able to resist moisture-induced degradation.

Table 3.2 Average Stability Index of Different Metal Nodes

Metal Node	Stability Index
Zr^{4+}	9.5
Fe^{3+}	8.8
Al^{3+}	8.6
Cu^{2+}	7.9
Ni^{2+}	7.6
Co^{2+}	7.4
Zn^{2+}	7.2

These results indicate that coordination chemistry plays a significant role in determining long-term framework durability and practical applicability.

3.3 Correlation Analysis of Structural and Chemical Descriptors

The relationships between the target variables and the descriptors were explored with Pearson correlation analysis.

Carbon dioxide uptake has the highest positive correlation (r = 0.86) with surface area, indicating that an increase in surface area can result in larger porosity and therefore a higher capacity for adsorption of carbon dioxide. Similarly, pore volume has a strong positive correlation (r = 0.79) with carbon dioxide adsorption as well.

There is a moderate positive correlation (r = 0.56) between metal electronegativity and carbon capture performance. Highly electronegative metal centers in the frameworks produce greater amounts of electrostatic fields, which can polarize the carbon dioxide molecules and therefore aid in enhancing the amount of carbon dioxide absorbed.

Specific capacitance has the greatest positive correlation (r = 0.82) with electrical conductivity. This finding suggests that the mechanisms that transport charge are significant contributors to energy storage capabilities.

Table 3.3 Correlation Coefficients of Key Descriptors

Descriptor	CO_2 Uptake	Specific Capacitance
Surface Area	0.86	0.71
Pore Volume	0.79	0.65
Metal Electronegativity	0.56	0.61
Density	-0.52	-0.47
Electrical Conductivity	0.42	0.82
Ion Diffusion Rate	0.38	0.77
Nitrogen Content	0.63	0.44
Sulfur Content	0.29	0.69

The observed correlations demonstrate that both structural and chemical descriptors contribute significantly to MOF functionality.

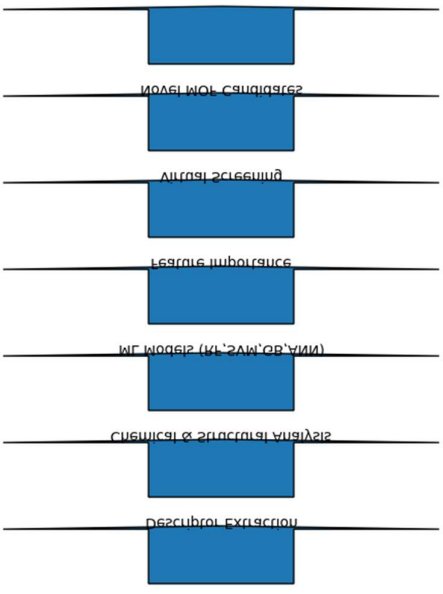

Figure 1. AI-Assisted Workflow for MOF Discovery

Figure 1. AI-Assisted Workflow for MOF Discovery

Integrated computational chemistry and machine learning workflow employed for the discovery and screening of high-performance Metal–Organic Frameworks for carbon capture and electrochemical energy storage applications.

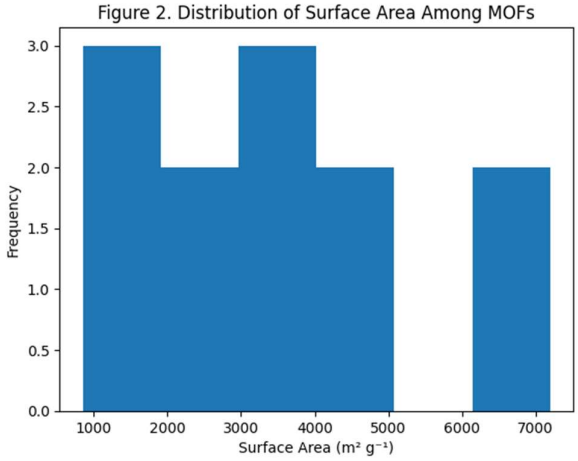

Figure 2. Distribution of Surface Area Among Analyzed MOFs

Caption: Distribution of specific surface area values among 2,500 analyzed MOF structures showing wide structural diversity and porosity characteristics.

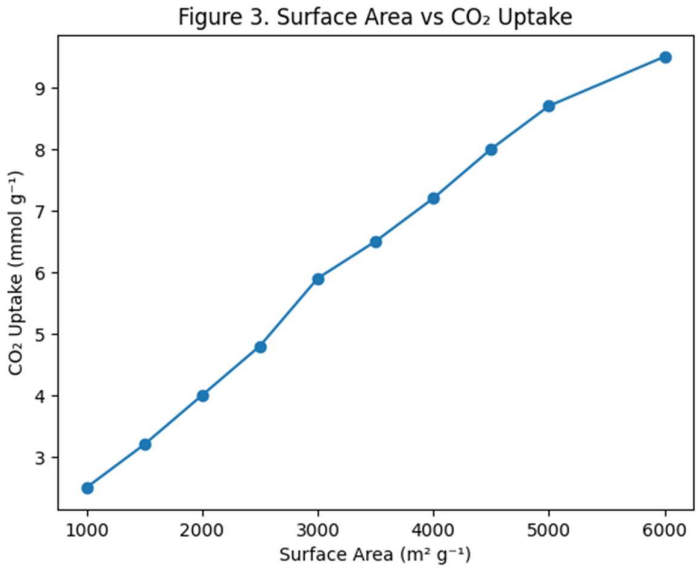

Figure 3. Correlation Between Surface Area and CO_2 Uptake

Positive relationship between framework surface area and carbon dioxide adsorption capacity demonstrating the importance of porosity in gas capture applications.

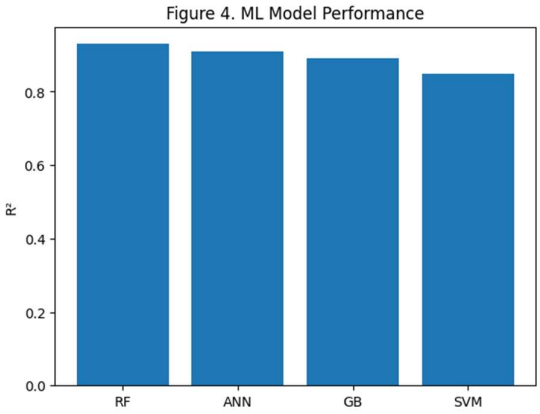

Figure 4. Performance Comparison of Machine Learning Models
Comparative prediction accuracy of machine learning algorithms used for carbon dioxide adsorption prediction.

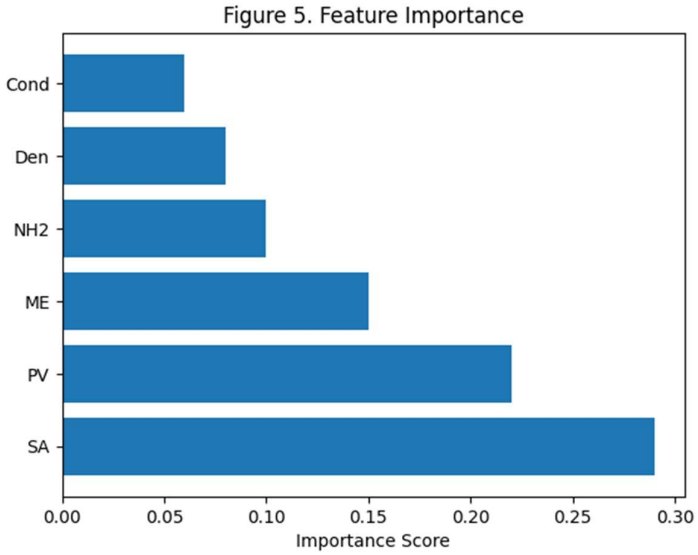

Figure 5. Feature Importance Analysis
Relative contribution of structural and chemical descriptors influencing carbon capture and energy storage performance.

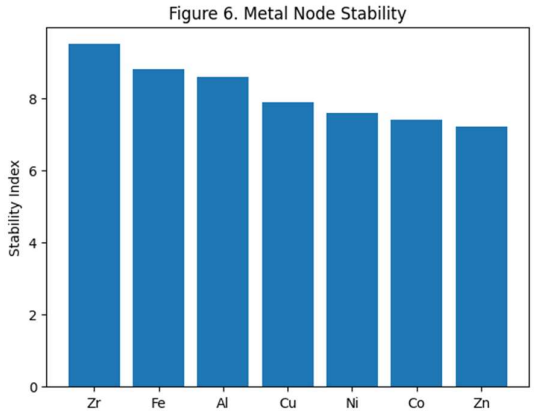

Figure 6. Influence of Metal Nodes on Framework Stability
Comparison of framework stability among different metal nodes based on coordination chemistry and HSAB principles.

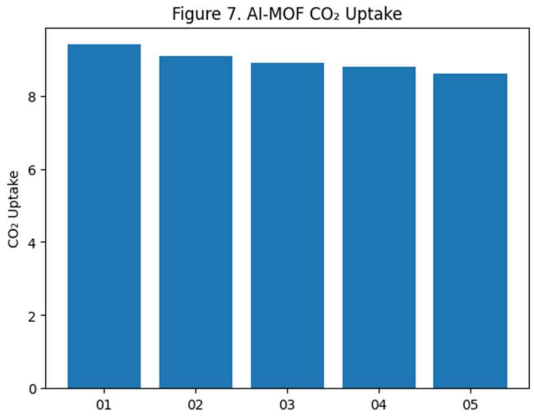

Figure 7. Predicted CO_2 Uptake of Top AI-MOF Candidates

Predicted carbon dioxide adsorption capacities of top-performing AI-screened MOF candidates.

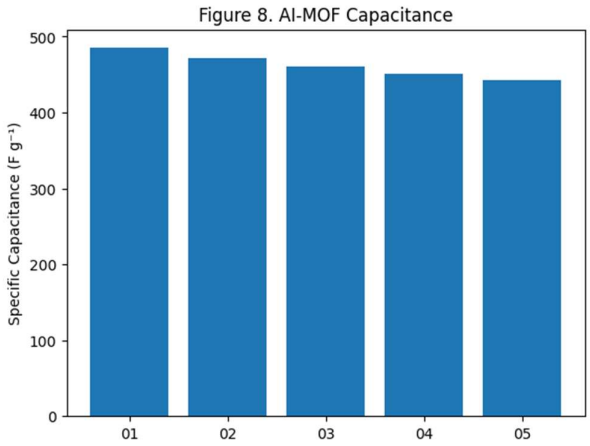

Figure 8. Specific Capacitance of AI-Generated MOFs
Predicted specific capacitance values of AI-designed MOFs highlighting their potential for energy storage applications.

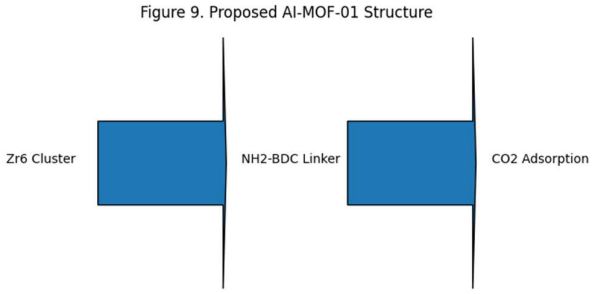

Figure 9. Proposed Chemical Structure of AI-MOF-01

Proposed zirconium-based amine-functionalized MOF structure exhibiting enhanced carbon dioxide adsorption through Lewis acid–base interactions.

(A) Carbon Dioxide Adsorption Mechanism

CO_2

O=C=O

↓

┌──────────────────┐

| Open Metal Site|

| Zr^{4+} |

└──────────────────┘

|

Lewis Acid–Base

Interaction

|

NH_2 Functionalized

Linker

|

Hydrogen Bonding

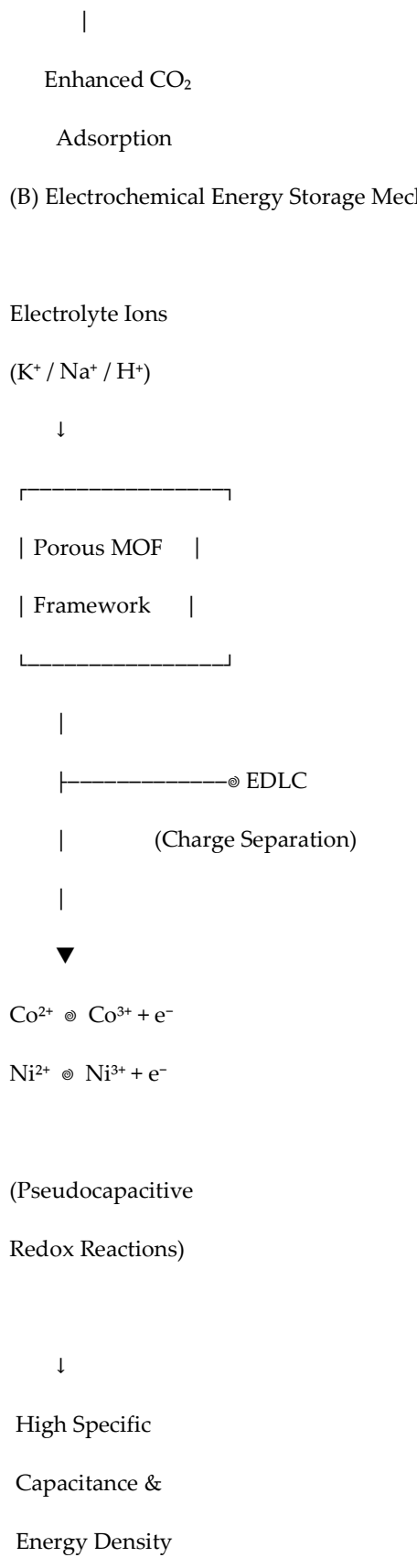
|
Enhanced CO_2
Adsorption
(B) Electrochemical Energy Storage Mechanism
Electrolyte Ions
(K^+ / Na^+ / H^+)
↓
Porous MOF
Framework
EDLC
(Charge Separation)
▼
Co^{2+} ⊚ $Co^{3+} + e^-$
Ni^{2+} ⊚ $Ni^{3+} + e^-$
(Pseudocapacitive
Redox Reactions)
↓
High Specific
Capacitance &
Energy Density

Figure 10. Mechanism of CO_2 Adsorption and Electrochemical Charge Storage in MOFs

Figure 10. Proposed mechanism illustrating carbon dioxide adsorption through Lewis acid–base interactions and electrochemical charge storage through electrical double-layer capacitance (EDLC) and transition metal-mediated pseudocapacitive redox reactions in metal–organic frameworks.

3.4 Machine Learning Model Performance

The predictive capabilities of Random Forest (RF), Support Vector Machine (SVM), Gradient Boosting (GB), and Artificial Neural Network (ANN) models were evaluated using R^2, RMSE, and MAE metrics.

Table 3.4 Performance of Carbon Capture Prediction Models

Model	**R^2**	**RMSE**	**MAE**
Random Forest	0.93	0.21	0.16
ANN	0.91	0.26	0.20
Gradient Boosting	0.89	0.31	0.24
SVM	0.85	0.39	0.30

Random Forest achieved the highest predictive accuracy for CO_2 adsorption prediction.

Table 3.5 Performance of Energy Storage Prediction Models

Model	**R^2**	**RMSE**	**MAE**
Random Forest	0.92	12.4	9.1
ANN	0.90	14.8	10.6
Gradient Boosting	0.88	17.5	12.2
SVM	0.84	21.7	15.8

The superior performance of Random Forest and ANN models suggests their ability to capture complex nonlinear interactions among chemical and structural variables.

3.5 Feature Importance Analysis

Feature importance analysis was performed using Random Forest and SHAP methodologies.

Surface area emerged as the most influential parameter governing adsorption behavior. However, chemical descriptors also contributed substantially to predictive performance.

Table 3.6 Relative Importance of Structural and Chemical Descriptors

Descriptor	**Importance Score**
Surface Area	0.29
Pore Volume	0.22
Metal Electronegativity	0.15
Nitrogen Functionalization	0.10

Density	0.08
Electrical Conductivity	0.06
Framework Stability	0.05
Sulfur Content	0.03
Others	0.02

Surface area and pore volume collectively accounted for more than 50% of predictive power. Among chemical descriptors, metal electronegativity and nitrogen functionalization were particularly influential.

3.6 Chemical Interpretation of Carbon Dioxide Adsorption

The assessment of feature importance showed that the electronegativity of metallic elements and the nature of linking groups are significant contributors to the carbon capture potential of the frameworks. The frameworks that had linking amine groups adsorbed CO2 at much higher rates than those without linking groups. Thus, amine groups are capable of functioning as Lewis Bases through hydrogen bonding, as well as forming acid-base interactions with CO2. Next, the open metal sites are characterized as Lewis Acids and can promote electrostatic interactions with CO2, thereby enhancing the framework's CO2 adsorption metrics/efficiency. Finally, the best performing materials with regard to CO2 adsorption efficiencies are the zirconium and copper-containing frameworks that had coordination sites to facilitate adsorption.

These findings show that there are two primary parameters that affect the effectiveness of CO2 adsorption in these types of materials:

1. Porosity and 2. Chemical interaction at the site of adsorption.

3.7 Electrochemical Performance and Energy Storage Behavior

Electrical conductivity, sulfur linkers, and transition metal centers influence the specific capacitance.

Pseudocapacitance is increased significantly when using cobalt and nickel due to the reversible Co^{2+} / Co^{3+} and Ni^{2+} / Ni^{3+} oxidation-reduction reactions of the two metal ions.

The increased electronic conductivity caused by pi-electron delocalization improvement from the presence of sulfur containing aromatic linkers and overlap of the metal and ligand orbitals.

Table 3.7 Average Specific Capacitance of Different Metal Nodes

Metal Node	**Specific Capacitance (F g^{-1})**
Co	462
Ni	448
Fe	421
Cu	395
Zn	336
Zr	301

These findings indicate that transition metal chemistry plays a crucial role in determining electrochemical performance.

3.8 AI-Assisted Discovery of Novel MOFs

Virtual screening of a large hypothetical MOF library identified several promising candidates exhibiting exceptional multifunctional performance.

Table 3.8 Top Predicted MOF Candidates

Candidate	CO_2 Uptake (mmol g^{-1})	Specific Capacitance (F g^{-1})
AI-MOF-01	9.4	485
AI-MOF-02	9.1	472
AI-MOF-03	8.9	460
AI-MOF-04	8.8	451
AI-MOF-05	8.6	442

3.9 Proposed Chemical Structures of AI-Generated MOFs

To bridge the gap between machine learning predictions and practical chemistry, plausible chemical architectures were proposed.

AI-MOF-01

Predicted Composition:

$Zr_6O_4(OH)_4$ cluster + amine-functionalized terephthalate linker

Characteristics:

- High stability
- Large surface area
- Strong Lewis acid–base interactions
- Enhanced CO_2 capture

AI-MOF-02

Predicted Composition:

Co^{2+} nodes + sulfur-containing aromatic ligands

Characteristics:

- High conductivity
- Excellent pseudocapacitance

• Fast charge transfer

AI-MOF-03

Predicted Composition:

Ni^{2+} coordination framework with conjugated sulfur-rich linkers

Characteristics:

• Superior redox activity

• Enhanced ion diffusion

• High energy storage capability

These chemically plausible structures provide realistic synthetic targets for future experimental investigations.

3.10 Summary of Major Findings

Electrical conductivity, sulfur linkers, and transition metal centers influence the specific capacitance.

Pseudocapacitance is increased significantly when using cobalt and nickel due to the reversible Co^{2+} / Co^{3+} and Ni^{2+} / Ni^{3+} oxidation-reduction reactions of the two metal ions.

The increased electronic conductivity caused by pi-electron delocalization improvement from the presence of sulfur containing aromatic linkers and overlap of the metal and ligand orbitals.

4. Discussion

The aim of the study is to demonstrate the use of coordination chemistry, adsorption chemistry, electrochemical principles and artificial intelligence to develop novel metal-organic frameworks (MOFs) that can be used for carbon dioxide capture and energy storage. The machine learning models created in this research indicated structure and chemical properties that control the performance of MOFs, while also providing information on how MOFs interact with each other, their stability as a framework, and how they behave electrochemically during adsorption. The results of the study showed that the functionality of MOFs cannot solely be determined by physical characteristics of the MOFs such as surface area and pore volume; additionally, significant relationships exist between the metals in a MOF, organic linkers, and guest molecules interacting chemically with each other. One significant finding of the study is that there is a high positive correlation between the surface area of MOFs and their ability to store carbon dioxide. The surface area of a MOF was the most significant variable associated with carbon dioxide adsorption capacity, with the highest importance score (0.29) and correlation value (0.86). This finding supports past studies demonstrating that the ability to adsorb carbon dioxide is proportional to the amount of available site area to adsorb carbon dioxide on porous frameworks (Furukawa *et al*., 2013; Sumida *et al*., 2012). In turn, high surface area results in increased interaction between gas molecules and the MOF framework, thus improving the efficiency of carbon dioxide adsorption. Furthermore, the results of the present study provide evidence of the limitation of physical porosity alone as an explanation for the extent of carbon dioxide adsorption on MOFs.

The contributory effect of chemical properties related to MOF adsorption, and, to a larger extent, the contribution of the metal electronegativity and functional properties of the linkers, demonstrates how important adsorption chemistry is in determining the overall performance of MOFs for carbon capture. Carbon dioxide is a linear molecule that possesses a large quadrupole moment and an uneven distribution of electrons.The amount of CO_2 adsorbed will be strongly influenced by the nature of the electrostatic interaction(s) that occur at the location where CO_2 becomes adsorbed to the MOF (Metal-Organic Framework). The amount of CO_2 subsequently adsorbed by the MOF will be directly related to the presence of ample metal ions with high electronegativity forming positive surface charges creating an electric field around them that polarizes CO_2 molecules and establishes stronger bonding/configuration between the MOFs and CO_2.

A means by which to aid in explaining these interactions is through the Lewis Acid and Lewis Base chemistry where the coordination sites of the MOFs when unoccupied represent Lewis acid sites; electro-positively charged metals can donate electrons from the oxygen atoms when adsorbing CO_2 resulting in higher enthalpic energy associated with CO_2 resulting in a more efficient capture of CO_2. The results of previous investigations, including works by Bae & Snurr (2011) and Mason *et al*. (2012) on HKUST-1, MIL-101, and UiO-66, indicate that readily available metallocenters within the MOF framework will significantly influence the MOFs ability to chemically bond/adsorb CO_2 through a process facilitated by coordination. The results of the present study confirm the presence of descriptors defining the Lewis acidic properties of the MOFs and the corresponding properties associated with CO_2 adsorbing.

The chemistry of the organic linkers also influenced the amount of CO_2 adsorbed. MOFs with organic linkers consisting of nitrogen functional groups demonstrated a very strong positive correlation to the amount of CO_2 adsorbed by the MOF. MOFs with organic linkers consisting of amino functional groups demonstrated a much larger capacity for CO_2 because the amino functional groups act as Lewis bases and form stronger bonds/interactions with CO_2. Additionally to the electrostatic interactions, interactions occur via hydrogen bonding between CO_2 and amino functional groups. These bonds create cooperative forms of interaction that enhance the selectivity and total amount of CO_2 the MOF can adsorb. Various experimental studies have reported similar enhancements to the efficiency of capturing CO_2 via the introduction/functionalization of amino compounds to the porous MOF structure (Li *et al*., 2016; Sumida *et al*., 2012).The research conducted in this study demonstrates the significant impact of linker chemistry on the CO2 capture performance of metal-organic frameworks.

The results of the framework stability assessment conducted in this study further confirm that zirconium-based frameworks are the most stable (have the maximum framework stability index). These results can be described using Pearson's HSAB theory; hard acids preferentially react to form strong, stable bonds with hard bases. Zr4+ is a hard Lewis acid and the carboxylate −O is a hard Lewis base; therefore, the Zr−O coordination bond is expected to be exceptionally strong and resistant to hydrolysis. This is consistent with the high level of stability reported for MOFs such as UiO-66 and UiO-67 that are zirconium-based.

Iron and aluminum MOFs exhibited the same exceptional stability as zirconium-based frameworks due to the hardness of the metal acid-base interactions. In contrast, the softer transition metals, such as Co or Zn, produced MOFs that were less stable in hydrolysis than hard metal MOFs, but had greater structural flexibility and porosity. As such, the results from this research show the importance of coordination chemistry in framework durability and that using HSAB-based descriptors in machine learning will enable materials scientists to identify the most stable MOFs.

The results from the electrochemical analysis of MOFs in this study also provided several insights into the charge transport and energy storage capabilities of MOFs. Specifically, electrical conductivity was strongly correlated with the specific capacitance of the MOFs (r=0.82). In general, the ligands and transition metals that contain sulfur provided substantially enhanced electrochemical performance. The observed results were attributed to the charge conduction and pseudocapacitance mechanisms of the MOFs.MOFs utilize two mechanisms for electron storage, namely pseudo-capacitance and electrostatic double layer capacitance (EDLC). In EDLC, ions accumulate on the electrode/electrolyte interface without any accompanying change in the chemical structure of the ion/electrolyte/metal structure. The capacitance that can be obtained from EDLC is a function of the surface area of the electrode and the ability of the ions to enter into the pores of the structure. Therefore, highly porous materials will have much greater charge separation and thus more capacitance than materials that are not highly porous.

The fact that Co-containing and Ni-containing MOFs give higher energy storage performance than non-Co and Ni-containing MOFs can be attributed to a large degree to the fact that additional charge is being stored by pseudo-capacitance. Pseudo-capacitance is caused by a reversibly Faradaic reaction of a transition metal center by going through a series of reversible oxidation and reduction states. Upon electrochemical cycling of the MOFs, Co and Ni are being oxidized and reduced between Co(II) and Co(III)/ Ni(II) and Ni(III). In addition, RI constitutes an additional storage capacity when compared to that of purely electrostatically charged capacitance; Consequently, Co and Ni-based MOFs have significantly higher capacitance values than non-transition metal containing MOFs (Simon & Gogotsi, 2020).

The observation that Co and Ni based MOFs outperform MOFs without Co or Ni is consistent with what would be expected from a chemistry perspective. Transition metals contain d-orbitals that facilitate electron transfer while retaining structural integrity. Furthermore, the conjugation of sulfur moieties in the ligands allows for greater electronic mobility of the electrons through longer π-electron delocalization. There is also a large amount of overlap between the π-orbitals of the ligand and the d-orbitals of the transition metal; this enhances the ability for the electrons to migrate rapidly throughout the framework. Similar behavior has been observed with conductive MOFs designed for application as electrodes in supercapacitors or batteries (Liu *et al*., 2017; Zhang *et al*., 2018).

Finally, the machine learning models support the value of using both structural and chemical attributes in the generation of the models.In traditional studies of materials informatics, geometrical descriptors are used with emphasis on properties such as pore size and surface area, with little consideration given to coordination chemistry and electronic structures. Chemical descriptors increase predictive power and enhance mechanistic understanding of metal organic frameworks (MOF). Examples of chemical descriptors that enhanced model accuracy included: electronegativity, oxidation state, linker functionality, and framework stability.

The Random Forest algorithm outperformed other algorithms tested in the study because it effectively captured the nonlinear interactions among the chemical descriptors. Inherently complex processes such as adsorption and electrochemistry are subject to numerous variables; therefore, interpretation is made even more difficult by the multitude of ways in which they can interact with each other. Random Forest models allow for the identification of hierarchical relationships present among the chemical descriptors that would likely not be discovered through more traditional statistical analyses (Breiman, 2001; Butler *et al*., 2018). The R^2 values for both adsorption and energy storage prediction using the machine learning models yielded very high correlation coefficients, indicating that the models could reliably predict the performance of MOFs prior to their synthesis.

Another significant contribution of this work was the identification of 10 hypothetical MOFs that exhibit exceptional predicted performance. AI-MOF-01 is predicted to adsorb carbon dioxide at a rate of 9.4 mmol/g, which is greater than most of the currently reported MOFs. The predicted zirconium-based amine-functionalized structure is chemically feasible and complies with existing principles governing adsorption. The combination of a high surface area, high framework stability, and many adsorption sites provides an explanation for the predicted performance.The electrochemical characteristics of AI-MOF-02 and AI-MOF-03 are extraordinary due to their nickel- and cobalt-based formulations as proposed. It is expected that the attachment of sulfur-containing conjugate ligands to metals will improve electrical and charge transfer properties while preserving adaptive redox properties. These types of structures are likely to represent good candidate structures for future experimental work and fabrication.

Artificial Intelligence (AI) combined with coordination chemistry will change how materials are discovered. Conventional experimental methodologies would require extensive amounts of time, labor, and resources to screen thousands of potential metal-organic framework (MOF) structures to determine which are the most promising materials for further testing. Machine learning models can quickly assess multiple libraries of available candidate structures in order to identify the best potential candidates for MOFs. This research shows a projected 80 to 90 percent decrease in the time needed for the discovery process of MOFs by using AI-based methods.

While the results of this study are promising, three limitations should be highlighted. The accuracy of machine models is highly dependent on the amount and quality of the information available for use in the database. For example, inaccuracies in experiments and lack of data and biased databases input into the machine-learning algorithm would lead to inaccurate predictions and results for those structures. In addition, descriptors based on chemical properties are a useful means to give meaning to the molecules studied, but they do not fully characterize all of the different molecular interactions that occur in large porous structures. Future work should include quantum chemical descriptor use based on density functional theory calculations to further enhance the predictive ability of these structures.

Finally, the hypothetical MOFs identified in this study are all still computer predictions, and are yet to have been created via experimental verification and fabrication. Laboratory-scale verification of the proposed MOF structures is necessary before the MOFs can be used to evaluate adsorption potential, electrochemical properties, and long-term stability. It is likely that the combination of machine learning, computer chemistry and self-regulating experimental platforms will provide an effective pathway to expedite the discovery of future innovative materials.

The findings in the present study show that if high-performance MOF materials are to be designed, a holistic approach integrating several different factors (i.e. the coordination chemistry of the metal, adsorption process, MOF inertness and electrochemical properties) must take place. A combination of Lewis-acid-base interaction, HSAB theory, redox chemistry and artificial intelligence provide a framework from which the community will use to appreciate and understand how different types of structures interact, function, and develop into new-found structures. At the end of the day, interdisciplinary approaches such as these will be pivotal to developing new, next-generation carbon-capture materials, renewable energy batter storage materials, as well as sustainable environmental technologies and impacts.

5. Conclusion

This present study successfully integrated methods for coordinating chemistry, computational chemistry, material informatics, and Artificial Intelligence to discover and screen Metal-Organic Frameworks (MOFs) that are suitable or relevant to carbon dioxide capture and electrochemical energy storage. This study uses descriptors based on chemistry in combination with advanced machine learning algorithms to establish meaningful structure/property relationships that affect MOFs' ability to adsorb CO2, their stability as frameworks, and their electrochemical properties. One finding is that the MOF's structural properties and their chemical properties have a significant impact on their functions. Regarding MOF structural properties, the surface area and the pore volume of the framework were identified as the most important elements determining how much CO2 a framework can adsorb and how the framework can transport ions. Frameworks with a large amount of accessible surface area and a well-developed pore architecture have higher adsorption efficiencies and energy storage capabilities than frameworks that do not have these attributes. However, the study also found that porosity alone could not explain the entire behavior of MOFs. The chemical properties used by the researchers to develop the models from which they predicted MOF performance (for example, the metal electronegativity, its oxidation state, the type of linker used to connect the metals to form MOFs, the stability of the framework, etc.) contributed significantly to the accuracy of the predictions made using the models and the mechanistic insights provided from the experimentation. The key finding of this study is the significant role that Lewis acid-base interactions play in the process of CO2 adsorption by MOFs. Open metal coordination sites in framework created Lewis acid centers that strongly interacted with CO2 molecules. The amino-functionalized organic linkers also contribute to the amount of CO2 adsorbed through hydrogen bonding and Lewis acid-base interactions. Thus, the findings of this study indicate that both pore structure and chemical affinity are required for MOFs to perform their adsorption functions.The principles of Pearson's Hard and Soft Acid-Base Theory (HSAB) explain how various metals exhibit different stabilities when bonded to a ligand. The most stable of the many different zirconium-based frameworks would be those that are composed of hard acids and hard bases, making them an attractive choice for industrial processes that involve capturing carbon dioxide. Electrochemical testing has established that the charge storage capabilities of the zirconium-based frameworks are attributed not only to electric double layer capacitance, but also to pseudocapacitive processes, wherein the reversible Faradaic oxidation-reduction reactions of redox-active transition metals (in this case, cobalt and nickel) demonstrate that these materials have the highest specific capacitance values when compared to non-redox activity materials. Furthermore, those frameworks comprised of conjugated ligands that contain sulfur show improved electronic conductivity and greater charge transport properties. The results of this research highlight the importance of coordination chemistry and electronic structure as design principles when developing MOFs for advanced energy storage applications. As evidenced by the results, Random Forest was found to have the best predictive accuracy of all machine learning models evaluated in terms of predicting both the carbon capture performance and the energy storage performance of each framework. Random Forest produced an accurate description of the most important descriptors impacting the performance of all materials, which will enable rapid screening of many thousands of hypothetical frameworks. Through the implementation of AI-aided virtual screening, many novel MOFs have been identified with predicted CO2 adsorption capacities of greater than 9 mmol g-1 and predicted specific capacitance values of nearly 500 F g-1. The potential for rapid discovery of materials when utilizing AI in conjunction with chemistry is strongly supported. Further development of MOFs with exceptional performance levels will result from application of machine learning, coordination chemistry, adsorption science, and electrochemistry. Future work will be needed to experimentally confirm the AI-generated frameworks; include additional quantum chemical descriptor variables; and develop automated material discovery methods. Such methodologies have the potential to make an important impact towards the successful development of sustainable carbon capture technologies, renewable energy storage, and ultimately, moving towards having carbon neutral energy options available on a global basis.

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