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Exploring covalent organic frameworks through the lens of computational chemistry

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Covalent organic frameworks (COFs) are a special class of porous polymeric architectures with robust covalent bonds that can have two- or three-dimensional frameworks. They can easily be utilised for designing primary and higher-order architectures, and their unique properties make them a groundbreaking class of materials in the current scientific research. The advantageous properties of COFs include easily tunable porous structures, high surface area, low density, and exceptional chemical and thermal stability. With their diverse molecular structures, they are promising candidates for ubiquitous applications including energy and the environment. Employing various advanced computational methods such as grand canonical Monte Carlo (GCMC) simulations, molecular dynamics (MD) simulations, density functional theory (DFT), hybrid quantum mechanical/molecular mechanics (QM/MM), multiscale modelling and machine learning (ML) approaches, researchers provided a profound understanding of the interaction/adsorption mechanisms, performance–structure relationships and electronic behaviour of COFs even at the atomic level, which is often unattainable through conventional experimental methods. Currently, emerging computational methodologies play more imperative roles in unveiling the exciting potential of COFs in mitigating global challenges pertaining to environment, energy and sustainability. This review explores the critical role of computational chemistry in the design, modelling, and applications of COFs, including key challenges and future perspectives, serving as a stepping stone for future advancements.

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1. Introduction

Covalent organic frameworks (COFs) are an attractive class of porous materials with exceptional physicochemical properties



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and fully predesignable nature.¹ The advantages of COFs include extensive surface area, easy modifiability, tunable pores, and exceptional mechanical, thermal and chemical stability.^{2,3} Nowadays, for the synthesis of COFs, trends have shifted from energy-intensive hydrothermal/solvothermal approaches to more energy-saving and environmentally benign synthetic procedures such as microwave irradiation, electron beam irradiation, and ultrasound irradiation.⁴ COFs came into focus at the beginning of 2000s and were synthesized based on the combined concept of organic chemistry synthesis and coordination chemistry. This synergistic approach was carried out to design novel materials having zeolite or MOF-type porous architectures, which also have the advantages of easy tunability as well as flexibility like organic polymers.⁵ Yaghi *et al.*, in 2005, designed the first 2D COFs (COF-1 and COF-5), where a boronate-ester linkage was used for the synthesis. They set the benchmark for the use of porous materials in material science research.⁶ Again in 2007, Yaghi and colleagues became the first to design 3D COFs (COF-102, COF-103, COF-105, and COF-108), employing co-condensation and self-condensation reactions of tetrahedral tetra(4-dihydroxyborylphenyl)methane (TBPM), silane analog of TBPM, *i.e.* TBPS, and triangular hexahydroxytriphenylene (HHTP). Among the designed COFs, COF-108 had the lowest density as compared to the other reported crystalline materials at that time.⁷ A pictorial representation of the advantageous properties of COFs and their broad range of applications is shown in Fig. 1.⁸ A survey on the number of publications on “covalent organic frameworks” between 2015 and 2025 was conducted, which is represented in Fig. 2. It is clearly observed that the demand for covalent organic frameworks has been increasing day by day. In the literature, there are approximately 500 000 different types of reported COFs, where the majority of them exhibit pore sizes ranging from 1 to 3 nm.⁹ With their adjustable porous nature,



Fig. 1 Diverse properties and applications of COFs.

extraordinary stability and high surface area, COFs are suitable for a wide range of separation- and adsorption-based applications such as sensing, catalysis, gas separation, and storage applications.^{10–12} Due to their high chemical and thermal stability, COFs can withstand harsh physicochemical conditions, which also makes them potential candidates for various industrial applications. To screen out the application-specific materials in computational COF research, the primarily used databases are (i) computation-ready and experimental COFs (CoRE COFs), (ii) clean, uniform, and refined with automatic tracking from experimental database COFs (CURATED COFs), and (iii) hypothetical COFs (hypoCOFs) database.¹³ While discussing COFs, their design, synthesis routes, structures and



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including chiral separation, biomolecule separation, and gas separation, wastewater treatment, membrane-based isolation and purification of natural products, biochemical reaction engineering and mechanism, kinetic analysis of reaction diffusion, modelling and simulation, and QSAR-QSPR studies.





Fig. 2 Publications on covalent organic frameworks in the last ten years (data were collected from the Web of Science employing the term “covalent organic framework”, accessed on 9th April, 2025).

functions are very important aspects to consider. A hierarchical diagram showcasing these important features of COFs and the corresponding research scopes is given in Fig. 3.

To understand the design conditions and principles of COF synthesis at the atomic scale, the computational approaches are highly effective, offering hazard-free, cost-efficient and safer alternatives to conventional methodologies that require expensive and hazardous chemicals.^{13,14} As already mentioned, COFs can have both 2D and 3D structures. Various reported 2D and 3D COFs based on their shapes/structures are shown in Fig. 4.¹⁵ Evaluating the exciting potential of COFs from the computational point of view, prior to carrying out experimental work, allows us to delve into the change in material's behaviour or properties at molecular/atomic levels (under various modifications). Herein, a holistic up-to-date review, mainly focusing on using various computational tools for screening the structure, optimization and property evaluation for numerous applications of COFs, is presented.

Owing to high surface area and tunable pore structures, COFs are extensively used in gas storage and separation applications, where computational studies can effectively boost their performance by exploring new possible designs. Ease of structural control and functionalization with meticulously arranged porous structure and enhanced selectivity make COFs attract tremendous interest from the scientific community. Computational screening of the building blocks and in-depth grasp of structural parameters as well as functional groups may lead to the design of novel COFs for a wide range of applications.¹⁶

Before synthesis, computational studies can provide predictions of topologies and structural stability of a COF framework. Density Functional Theory (DFT) and Molecular Dynamics (MD) simulations can reveal the stability, bonding and energetics of a COF architecture. Moreover, DFT and hybrid functionals can elucidate the electronic properties such as frontier orbitals, band gaps, and charge distributions in COFs, which are highly essential for designing effective COF-based tailored materials for their applications in sensing, energy harvesting,

and photocatalysis.¹⁷ Grand Canonical Monte Carlo (GCMC) and MD simulations provide insights into the adsorption, separation and diffusion behaviour of COFs, narrowing down the need for substantial experimental validation. From a huge database, computationally assisted screening can effectively pinpoint the most suitable COFs with superior (optimal) properties for particular applications.^{17,18} Non-equilibrium molecular dynamics (NEMD) simulations can provide an understanding of heat and mass transfer in COF membranes, including diffusivity, thermal conductivity and permeability.¹⁹ In-depth computational studies can explore charge transfer and storage in COFs and simulations of the integration of COFs into advanced battery systems, which may provide a new avenue for the development of next-generation batteries. Computational investigation of the electronic structure–property relationship may accelerate the design of COFs for green catalysis, environmental monitoring and biosensor applications. Computational interaction analysis of COFs with drugs and biomolecules can provide a pathway for their use in effective and controlled drug delivery applications.¹⁶ This account may inspire COF-based research and may introduce a new paradigm guiding the innovation of COF-based tailored materials with superior properties in the upcoming years. Furthermore, the study offers the researchers a nuanced understanding of the existing experimental and hypothetical COFs (with their property–performance relationship), and their implementation across diverse domains, providing strategies to design novel COFs for their future prospects in research and industrial applications.

2. Computational framework for COF research

2.1. Quantum mechanical approaches

Quantum mechanical (QM) approaches are powerful and widely used tools that leverage the concept of the uncertainty principles, quantum entanglement and dual nature of wave unveiling insights into the atomic and electronic behaviour within the COF frameworks.^{20,21} Density functional theory (DFT) and hybrid quantum mechanical or molecular mechanical approaches are the most widely employed QM methods for modelling as well as accurate prediction of various properties of COFs, as discussed in the following sections of this review.

2.1.1. Density functional theory (DFT). DFT is one of the most widely accepted, reliable and powerful computational tools to study many-body systems. Through precise calculations of total energy, electron density and the optimization of molecular structures, it offers an in-depth understanding of the electronic behaviour of complex systems such as COFs. Meanwhile, it figures out the most stable COF architecture for particular applications, with optimal precision (through energy optimization). Additionally, band gap study using frontier molecular orbital (FMO) analysis and conductivity analysis of COFs can also be carried out utilizing DFT. In order to optimize the geometrical structures and total energy calculations, the Kohn–Sham equation is employed. The Kohn–Sham equation



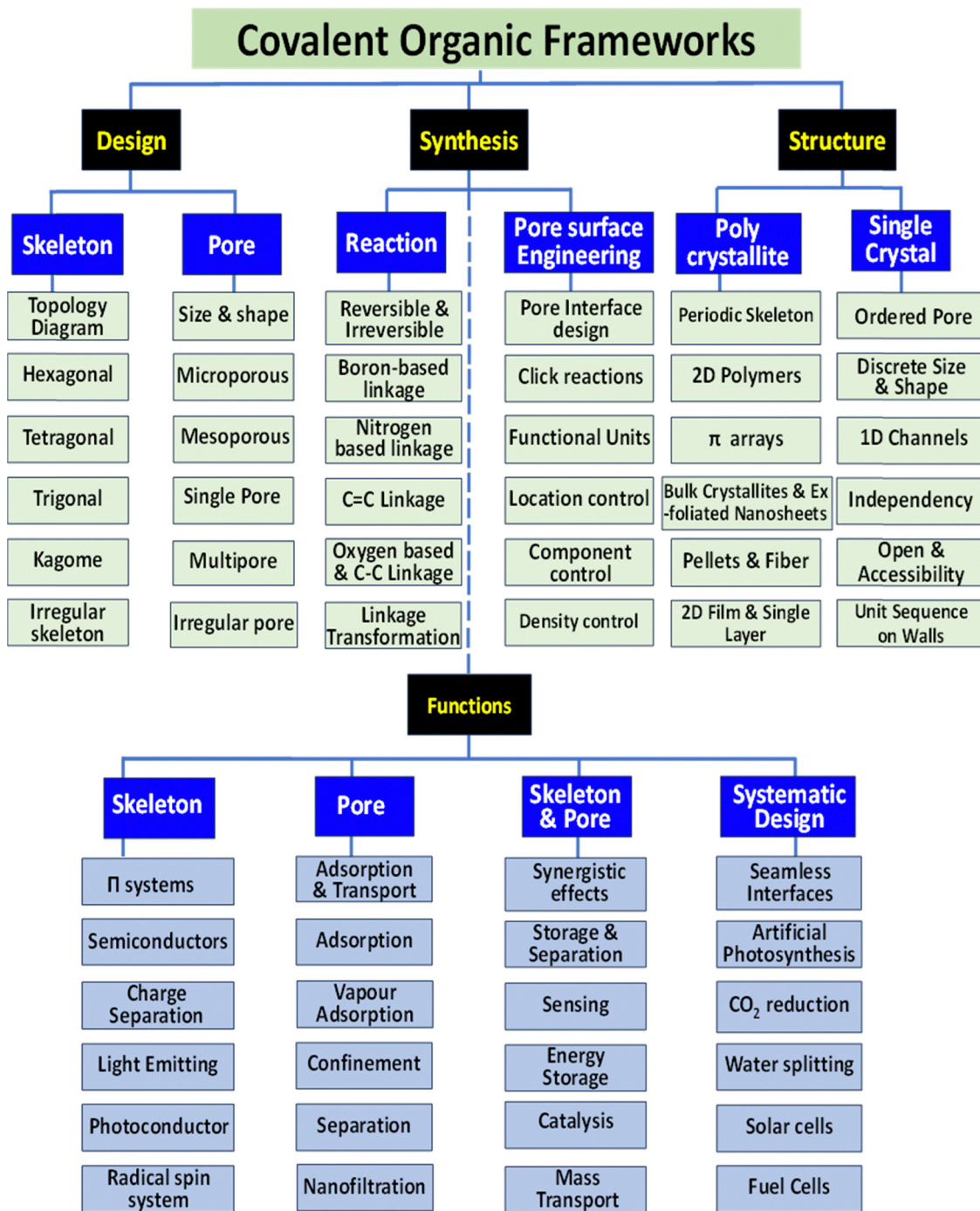


Fig. 3 Research scope of COFs in various domains. Reproduced from ref. 5 with permission from the Royal Society of Chemistry, Copyright 2021.

(eqn (1)) is given as follows:^{22,23}

$$\left[-\left(\frac{\hbar^2}{2m}\right)\nabla^2 + V_{(r)} + V_{H(r)} + V_{\{XC\}(r)} \right] \Psi_{i(r)} = \varepsilon_i \Psi_{i(r)} \quad (1)$$

where the wave function is denoted by $\Psi_{i(r)}$ that characterizes the dynamics of the i th electron, where ε_i denotes the associated energy. In the above expression, external potential, Hartree potential and exchange–correlation potential are indicated by $V_{(r)}$, $V_{H(r)}$ and $V_{\{XC\}(r)}$, respectively.

The external potential considers the repulsive potential of the nucleus with external field potentials and the Hartree potential demonstrates the electrostatic interaction of electrons. The electron correlation and electron exchange effects are taken into account by the exchange–correlation potential.

DFT models are used to calculate the adsorption/desorption energies of various systems using eqn (2):^{22,24}

$$E_{\text{adsorption/desorption}} = E_{\text{adsorbates}} + E_{\text{host}} - E_{\text{adsorption/host}} \quad (2)$$





Fig. 4 Various reported 2D (A) and 3D (B) COFs based on their shapes/structures. Reproduced from ref. 15 with permission from Elsevier, Copyright 2021.

where $E_{\text{adsorbates}}$, E_{host} and $E_{\text{adsorption/host}}$ denote the energies of the adsorbates, host and host-guest system, respectively.

BLYP and PBE are the most widely used generalized gradient approximation (GGA) used in DFT for the calculation of electron density. Even though they are commonly used, they have some limitations such as higher computational cost and may produce some errors. Hybrid DFT functionals, for instance, B3LYP and PBE0 can overcome these limitations up to a good extent, during the study of the electronic properties of

COFs.^{25–30} Experimental investigations integrated with DFT calculations and molecular dynamics (MD) simulations were reported for the exploration of electronic properties, layer interactions and charge carrier dynamics in benzodithiophene-COF thin films.³¹ From the findings, it can be concluded that the films with less thickness exhibit significantly higher hole mobilities (especially under light), indicating their potential in optoelectronics applications. In another study, an experimental study and DFT-based calculations were carried



out to evaluate the removal of naproxen (a pain-relieving medication), owing to its environmental concern, using an ionic guanidinium-based COF. The COF was found to exhibit excellent adsorption towards naproxen (308 mg g^{-1}), where physical adsorption was dominant over the chemical adsorption. The pi-pi and electrostatic interactions played an important role during the adsorption process, which was found to be exothermic and spontaneous. The guanidium-based COF architecture was reported as an effective and expeditious approach to mitigate the urgent issue of contamination of an aqueous stream by the naproxen drug (mainly after the COVID-19 pandemic). Meanwhile, the interaction of naproxen with the guanidium-based COF architecture (especially between the -COOH group of the naproxen molecules and the guanidinium of the COF) was thoroughly inspected, harnessing advanced computational techniques such as gradient model based on Hirshfeld partition and electrostatic potential analysis.³² The synthesis of fluorescent COFs, their fluorescence mechanism (analyzed employing DFT) and excellent heavy metal detection capabilities were envisioned in another work.³³ The intriguing impact of the intercalation of first-row transition metals into 3D COFs, their electronic properties, and structural changes revealing stable configurations was comprehensively investigated leveraging DFT calculations.³⁴ The study provided crucial insights into the structure-property relationships of transition-metal-intercalated COFs and enhanced the conductivity of COFs upon intercalation. Further, DFT studies, with the support of electron density mapping, provided insights into the structural properties, electrostatic properties and specific inter-layer interaction of 2D COFs.³⁵

The impact of protonation and structural variation on donor-acceptor COFs was also reported, revealing that upon protonation, the energy gap gets lowered and alters the electron dynamics within the COF framework, which was validated by experimental findings.³⁶ Notably, up to 98% of enantiomeric excess of racemic drugs was achieved by performing enantioselective separations using 3D chiral COFs (which are recyclable in nature), with tailored pore surfaces, and their chiral recognition mechanisms were elucidated with the aid of DFT approaches.³⁷

The integration of machine learning (ML) (specifically neural networks) has further revolutionized the field by improving and simplifying the classical DFT computations and boosting the calculation process. The use of classical DFT with the advantages of ML was discussed to analyze the gas adsorption behaviour of COFs following the Helmholtz energy model.³⁸

The semi-empirical approach uses experimental data to make the quantum mechanical calculation that reduces the computational cost.³⁹ By considering the core electrons as a part of the atomic nucleus, it omits the core electron integrals and only valence electrons are taken into account. AM1, PM3, PM6, OMx and NDO are the most frequently used semi-empirical methods, which are computationally way much faster than the *ab initio* methods, although there are certain trade-offs in accuracy in comparison to *ab initio* methods.^{39,40} A semi-

empirical method, termed the tight binding approach, combines the wavefunctions produced by individual/constituent atoms, to represent the wave functions of the whole system in order to strike a balance between accuracy with optimum speed (with an accuracy that is comparable to DFT).³⁹⁻⁴² DFTB is an extension of the tight-binding approach incorporated with advanced DFT principles. It uses precomputed interaction parameters, making the whole process significantly faster than traditional DFT. Despite the advantages, the dependence on various parameters for various elements and spin-polarization restricts their use on a broad scale.^{43,44}

2.1.2. Hybrid quantum mechanical/molecular mechanics (QM/MM) approaches. To study complex systems including COFs or modified COF-based architectures, QM/MM is considered as an emerging and powerful computational tool. This approach allows researchers to predict the molecular interaction and explore the electronic nature of massive systems while maintaining the computational cost.^{45,46} It seamlessly combines the quantum mechanical approach with molecular mechanics to fill the gap of the molecular mechanical (MM) approach with the quantum mechanical (QM) approach or vice versa. Using these approaches, quantum-level calculations can be achieved with the speed of classical mechanics (bridging the limitations of each method), for in-depth (with good precision) atomic-level calculations of molecular systems.⁴⁶⁻⁴⁸ The hybrid QM/MM approach utilizes quantum mechanical calculations for analysis of the focussed site of the system, while classical molecular mechanics evaluates the surrounding environment. An overall or detailed understanding of different massive systems like COFs can be achieved by employing this strategic synergistic approach, thereby effectively reducing the complexity of the calculation as well as the computational time. In this approach, during the computations of large/complex frameworks, additive and subtractive coupling pathways are often employed.^{47,48} The first pathway generally estimates the total energy obtained through the addition of the energies obtained by QM as well as MM approaches; therefore, it is termed as additive coupling. On the other way in case of the subtractive coupling, the entire system energy is calculated and then the QM component is added, considering the overlapping of QM as well as MM computations.⁴⁷⁻⁴⁹

In this approach, a complete system (CS) is partitioned into two parts, primary subsystem (PS) and secondary subsystem (SS), where PS is illustrated utilizing QM and SS is treated using the MM approach. The QM/MM energy of the complete system can be obtained by employing the following expression:^{47,48}

$$E(\text{QM/MM};\text{CS}) = E(\text{MM};\text{CS}) - E(\text{MM};\text{PS}) + E(\text{QM};\text{PS}) \quad (3)$$

where $E(\text{MM};\text{CS})$ and $E(\text{MM};\text{PS})$ denote the MM analysis of the complete system and the primary subsystem and $E(\text{QM};\text{PS})$ represents the QM analysis of the primary subsystem, respectively.

Even though the hybrid QM/MM approaches are crucial in biological and material science applications, in some cases the line of separation between QM and MM approaches can be tricky based on different systems. To overcome these



challenges, various strategies can be followed such as introducing linker atoms like hydrogen to cap the damaged bonds, regulating the effect of charges close to the borders, and modifying the borderline throughout the simulation. The versatility of the QM/MM approach was reported for studying the substituent effect on the enantioselectivity and reactivity of (salen)Mn catalysts within MOFs.⁴⁵ The crucial factors such as steric hindrance and electronic effects reducing enantioselectivity were portrayed in the study. Furthermore, the study depicted the impact of electronic and configurational changes on the catalytic performances of COFs, by adopting hybrid QM/MM approaches. Moreover, these methods can effectively explore the chemical reactions and interactions,⁴⁶ even at atomic/molecular levels, offering a new avenue of COFs or COF-based functional materials for their future advancements in a variety of domains, including drug design.

2.2. Classical force field methods

For the simulation of modelling purposes of various systems, classical force field methods primarily harness (predefined) potential energy functions to examine molecular behaviours under different conditions. These approaches can play a pivotal role in the thorough computational investigations of configurational/conformational properties, dynamic nature as well as in understanding numerous dynamic responses of COFs. This study presents various reported popular classical force field methods, analyzing the advantageous (electronic and structural) properties of numerous COFs and their potential applications with the ultimate goal of environmental remediations and sustainability.^{50–53}

2.2.1. Grand canonical Monte Carlo (GCMC) simulations.

GCMC simulations are pivotal in studying the real dynamics of systems by allowing the particles of the system to add, remove and move under energy considerations. Three distinct types of trial movements – formation, destruction and rotation/displacement of particles – are attempted while conducting GCMC simulations.⁵⁴ Through this approach, computer simulation is employed to understand the interaction/adsorption of gas molecules (or other various molecules) on the COF architecture with the variation in different parameters such as pressure and temperature (*i.e.* simulating a real-world environment).^{54–58} Moreover, the approach is suitable for the prediction/evaluation of adsorption isotherm, adsorption behaviour of various gases on different COF structures and selectivity parameter of different COFs towards some specific gases over other gases. Additionally, the GCMC approach is widely utilized by researchers to predict the impact of various functional sites, structures and pore sizes of the COFs on their efficiencies, providing valuable strategies to optimize these parameters.^{12,59–66} By optimizing these parameters of COFs, significant enhancement in selectivity, separation/adsorption, sensor applications, photovoltaic applications and photocatalytic activities were reported.^{67–70} This approach can be extensively used to understand the dynamic behaviour of the gas molecules within the porous COF framework/structure and for in-depth analysis of COF–gas interactions.

The algorithms mainly used by GCMC include the Metropolis and Gibbs algorithms. Metropolis algorithms are mainly employed for simulations of solid–liquid systems, whereas Gibbs algorithms are useful in the simulation of gas systems.^{22,71}

Employing the following expressions, the total adsorption value, for any system, can be defined:^{22,72,73}

$$N_{\text{total}} = N_{\text{excess}} + \rho V_{\text{free}} \quad (4)$$

where N_{total} and N_{excess} denote total and excess adsorption values, V_{free} denotes the single crystal cell's free volume, and ρ denotes the bulk density or packing density.

Again, the excess adsorption value can be calculated by employing the following equation:

$$N_{\text{excess}} = 1000 \left(\frac{N_{\text{am}} M_{\text{a}}}{N_{\text{a}}} - \rho V_{\text{free}} N_{\text{A}} \right) / (M_{\text{s}} M_{\text{a}}) \quad (5)$$

where N_{am} , M_{s} , M_{a} , N_{a} , and N_{A} represent the amount of adsorbate molecules (the final number), the quantity of materials of the single-crystal cell, the adsorbate's relative molecular mass, the number of structure cell and Avogadro's number, respectively.

Additionally, by harnessing GCMC simulations, the heat of adsorption (Q) can be calculated by solving the following equation:^{22,74}

$$Q = RT - \frac{\langle U_{\text{ff}} N_{\text{am}} \rangle - \langle U_{\text{ff}} \rangle \langle N_{\text{am}} \rangle}{\langle N_{\text{am}}^2 \rangle - \langle N_{\text{am}} \rangle \langle N_{\text{am}} \rangle} - \frac{\langle U_{\text{fs}} N_{\text{am}} \rangle - \langle U_{\text{fs}} \rangle \langle N_{\text{am}} \rangle}{\langle N_{\text{am}}^2 \rangle - \langle N_{\text{am}} \rangle \langle N_{\text{am}} \rangle} \quad (6)$$

where R is the gas constant, $\langle \rangle$ denotes the overall average, U_{ff} represents the potential energy of adsorbate molecules and the adsorbate–adsorbent potential energy is denoted by U_{fs} .

A larger value of Q indicates a strong affinity between the adsorbate and the adsorbent, thereby indicating a stable adsorption process between the two.

Upon integration with advanced and emerging technologies, including data mining and ML, the GCMC simulation becomes a robust and more powerful platform, paving the way for the synthesis of next-generation materials for gas storage/separations and environmental remediation. ML speeds up the evaluation process of complex systems, playing a crucial role in high-quality COF design strategies. The capabilities of COFs and MOFs, towards efficient energy storage, were compared by comparing their performances. Even though MOFs typically have a higher energy storage efficiency, the study highlights the potential of COFs to surpass MOFs in superior energy storage performance, through appropriate design strategies.⁷⁵

2.2.2. Molecular dynamics (MD) simulations. To gain crucial insights into the different systems, the MD simulation promotes computational investigations of atomic dynamics and interactions within the COF framework under different operating conditions.⁷⁶ The MD simulation guides the synthesis of defect-free and high-quality 2D COFs, by highlighting the optimization requirement of some key parameters such as temperature, speed of bond formation and surface



movement.⁷⁷ This approach explains how the 2D COFs can grow on a sheet of graphene, explaining the underlying mechanisms, emphasizing the significance of reaction temperature, van der Waals (vdW) interactions and monomer arrangements.⁷⁸ During MD simulations, the particles (atoms and molecules) follow Newton's equation of motion, which can be solved using integral iteration. The velocity and position coordinates of the particles at various moments can be obtained from the trajectory files of the simulation outputs.⁷⁹ In MD simulations, the trajectory of atoms can be obtained by employing Newton's equation of motion as follows:^{22,80}

$$\frac{d^2 l_i}{dt^2} = \frac{F_{l_i}}{m_i} \quad (7)$$

where the mass, position and force of the i th particle (in one direction) are denoted by m_i , l_i and F_{l_i} , respectively.

Additionally, the coefficient of self-diffusion in a system can be calculated using the following equation:^{22,81}

$$D = \frac{1}{6Nt} \sum_{i=1}^N \sum_{j=1, j \neq i}^N \langle (r_i(t) - r_j(t_0))^2 \rangle \quad (8)$$

In the above equation, $\langle \rangle$ denotes the overall average, and N , t , $r_i(t)$ and $r_j(t_0)$ denote the number of adsorbates, time of simulation, position of i th and j th molecule at time t and t_0 , respectively.

To tackle nanoplastic pollution, the role of COF was evaluated by Shang *et al.* using MD simulations.⁸² MD simulations were employed to study the adsorption behaviour of nanoplastics such as polyethylene (PE), polyethylene terephthalate (PET) and nylon-6 (PA 6) on functionalized COFs. It was observed that van der Waals interaction and electrostatic interaction play a key role in the adsorption process. Two-dimensional COF-1 and COF-5 were evaluated for their thermal conductivity using MD simulations, and it was found that they have very low thermal conductivity.^{83,84} A remarkable enhancement of thermal conductivity was observed upon the incorporation of C60 into COF-1, whose insight mechanism was also discussed in the study.⁸³ The findings have great importance in the practical use of COFs. For the first time, Liu *et al.* investigated the thermal conductivity/behaviour of 3D-boron-based COFs (COF-102, COF-103, COF-105, and COF-108) using MD simulations, which revealed their low thermal conductivity. The limited phonon mean free path was identified as the primary responsible factor for this.⁸⁵ The mechanical properties of the two-dimensional COFs (COF-1 and COF-5) and the effect of temperature on their elastic behaviour were evaluated using machine learning and MD simulation approaches.⁸⁶

To predict the transport of gas molecules through a porous framework, non-equilibrium MD (NEMD) simulations stand superior compared to equilibrium MD (EMD) simulations, as NEMD considers real transport effects such as the pore entrance and mass transfer resistances. NEMD simulations enable the investigation of gas flow behaviour through a nanoporous architecture, directly from the simulation itself, which can effectively consider pressure and concentration

gradient, demonstrate high accuracy and provide deeper insights.⁸⁷ In a study by Velioglu and Keskin, the GCMC + EMD approach was compared with the NEMD approach during the permeation study of CH₄ and H₂ through MOF membranes.¹⁹ It was observed that, as compared to the results obtained from the GCMC + EMD approach, the results from the NEMD approach showcased a close alignment with the experimental findings, highlighting the efficacy of the NEMD approach in revealing realistic gas transport mechanisms.

In a recently reported work, H₂ and CH₄ transport through COF membranes (COF-320 and COF-300) was evaluated leveraging the NEMD method. It was observed that the findings of the NEMD simulations closely match the experimental findings, as compared to the GCMC + EMD approach. The study also demonstrates the pivotal role of pore sizes for COF membrane's computational analysis and reveals the prominent COFs for the effective separation of CH₄ and H₂. As the diffusion and adsorption characteristics of the COF membranes are influenced by their pore sizes, the selectivity and permeation of the gases are highly based on their pore size range. The study showcased the importance of a simulation method for such studies, highlighting the robustness of the CGD-MD approach (gradient-driven molecular dynamics, a NEMD approach) for its reliable results with minimal error profiles.⁸⁷ In another study, the significance of the hydrophilicity of COF membranes during the permeation of Mg²⁺ and Li⁺ was investigated leveraging NEMD simulations. The study demonstrated that the membrane can effectively trap/adsorb Li⁺, while allowing Mg²⁺ to pass through it. The study may guide the fabrication of nanofiltration membranes for effective lithium extraction (or other ion sieving applications), which has the potential to meet the requirement for lithium in new energy sectors.⁸⁸ Wei *et al.*, in a recently reported work, have demonstrated the influence of functional groups and pore sizes on the effective separation of salt, employing NEMD simulations. The COF membrane TpPa-SO₃H displayed more than 98% rejection of Na₂SO₄, rivaling commercial membranes. The study may pave the way for next-generation membrane design, employing a simulation-based approach, without any experimental setup.⁸⁹

2.2.3. Machine learning (ML) approaches. As an emerging transformative paradigm and a powerful tool, ML prominently screens COFs that possess optimal efficiency across various domains. By leveraging specific algorithms, ML can efficiently figure out the potential candidates for a particular application, from a huge database, obtained from simulations like GCMC or MD.^{90–92} By employing the XGB model of ML (combined with simulation), the SF₆ and N₂ separation performance by COF-638 was investigated, underscoring the significance of porosity and adsorption heat.⁹³ The critical role of COF's building blocks and the impact of appropriate solvent selection during the synthesis of defect-free and high-quality COFs were demonstrated using ML approaches.⁹⁴ The automated ML approach outperforming traditional methods was reported for the prediction of the methane storage capacity of 403 959 COFs.⁹⁵



The integration of ML and first-principles calculations was reported for the prediction of the band-edge of 2D COFs (achieving high accuracy) in a fast and efficient manner.⁹⁶ In a recently reported work, ML facilitated the development of iron porphyrin COFs possessing enzymatic activity for the detection and identification of thiols, showcasing their potential in disease diagnosis applications.⁹⁷ The movement of protons in COFs was investigated using ML and MD simulations, revealing that the fastest movement was found in the pores, which may vary based on the side chain length, sulphonic acid concentration, and the water content.⁹⁸ It was seen that by reducing the side chain length and sulphonic acid concentration, while increasing the water levels, the proton mobility was significantly enhanced, demonstrating the potential of COFs in fuel cell applications. Moreover, ML and first-principles calculations were carried out for 2D porphyrin COFs to analyze their potential as thermoelectric candidates, and it was observed that the incorporation of metal atoms enhanced the stability and reduced the thermal conductivity, thereby improving thermoelectric performance.⁹⁹ Another study employed ML and first-principles calculations to provide a strategy for the predictions of band gap positions in 2D COFs, more precisely,^{96,100} facilitating advanced COF design strategies for their applications in nanoelectronics as well as in photocatalysis.

AI and ML-based advanced deep learning models (such as transformer-based models, convolutional neural networks, and graph neural networks) are employed by the researchers to gather various COF's structural parameters, including the presence of functional groups, surface area, and pore sizes, from a huge database, which is also termed parameter loading. Reliable parameter loading from open databases (for instance, CoRE-COF), employing automated tools such as molecular-level simulation and Python-based libraries, enhances the accuracy of the model. Specifically, physical, chemical and geometrical properties are the key parameters for this type of study, where the automated tools save time, reduce human effort and boost the material screening throughput. The data for a known COF obtained from a database are collected with their associated properties. From this, the model learns the pattern of chemical and structural features, enabling the system to understand the insightful structure–property relationship, a process known as model training. The model was then subjected to screening of huge databases to find the best-performing COFs with quantitative prediction of the desired properties, in a rapid manner. Yang *et al.* reported the efficiency of automated ML (AutoML) as compared to other conventional ML approaches, to analyse/predict the methane uptake capability of various COFs.⁹⁵ The study evaluated 403 959 COFs depending on their various chemical and physical properties. Furthermore, the study compared various conventional ML tools such as random forest, support vector machine, decision tree and multiple linear regression using the AutoML method Tree-based pipeline optimization tool (TPOT). The study highlights the potential of TPOT in terms of accuracy and speed that does not require any manual tuning, demonstrating superior prediction capability

and precision, making the approach more convenient and effective for COF research. In another work, the attention-based ML approach was employed to predict the thermal conductivity of COFs with high accuracy, emphasizing the role of dangling molecular branches. The transformer-based deep learning model (incorporated with the multi-head attention mechanism) was found to be more effective than the conventional ensemble-based regression approaches.¹⁰¹ The study evaluated 2471 two-dimensional COFs, and it was observed that COFs containing more dangling atoms have lower heat transfer capability owing to the disrupted pathways, which was further validated by vibrational analysis. The study showcased the potential of advanced deep learning methods to reveal the structure–property correlation of COFs, guiding researchers for quick and precise design of next-generation tailored COFs or COF-based materials, with desired properties.

2.3. Multiscale modelling techniques

The multiscale modelling techniques promote integrating diverse computational methods from multiple domains to address one specific issue.^{102–105} A study leverages a multiscale modelling approach to demonstrate the formation of 2D COFs at the interface of water and air.¹⁰⁵ The study involved an experimental study validated by an *in silico* approach. To achieve COFs with superior quality, preferential alignment of the building blocks at the interface, which promotes the polymerization process, was discussed. Through electronic and structural evaluations, the tunable band gap and semiconducting behaviour of the 2D COFs were substantiated, rendering them highly suitable for electronic applications. In tandem approach of various computational methods was reported to analyse the high efficacy of COFs in H₂ storage applications.^{106,107} Again, advanced quantum mechanical and classical force field methods were utilized for gaining an in-depth understanding of boron-based COFs, including their structural, thermal, mechanical and adsorption behaviours.¹⁰⁸

3. Applications of computational studies on COFs

3.1. Gas separation and storage

To study the adsorption behaviour of various gases on COFs, grand canonical Monte Carlo (GCMC) simulations are mostly employed. Owing to the flexibility of permitting variation in the total number of particles for adsorption analysis, it serves as a highly useful and frequently utilized tool for studying gas separation and storage applications.^{54–58}

Many COFs possess superior efficiency in gas separation applications in comparison to conventional porous materials like MOFs or Zeolites,^{109,110} which is established by computational evaluation. The exceptional performance of COFs towards the separation of industrial gas mixtures such as He/N₂, He/CH₄, CO₂/CH₄, CO₂/H₂ and CH₄/H₂ was observed, making them potential candidates in industrial gas separation and carbon capture applications. Table 1 shows the reported



Table 1 Computational approaches for COFs or COF-based materials, investigating their gas separation and storage potential with structural and dynamic analyses

Topic of study	COF/COF-based materials studied	Computational methods & algorithms	Studied parameters	Structural/dynamic testing	Ref.
Interaction of N ₂ and CO ₂ with COF	Nitrogen-containing COFs	<i>Ab initio</i> and DFT approach	Binding energy	Structural analysis	114
Separation of CO ₂ & H ₂ S from natural gas	580 synthesized and 891 hypoCOFs	DFT, GCMC simulations, multi-level computational screening	Separation, selectivity, structure–property correlation	Structural and dynamic analysis	13
Separation of CH ₄ /H ₂ , CH ₄ /N ₂ and C ₂ H ₆ /CH ₄ binary mixtures with 572 COFs	572 COFs	DFT calculations, MD and GC simulations, nucleus-independent chemical shift analysis	Adsorption, separation, binding energy	Structural and dynamic analysis	115
Adsorption of gases (<i>n</i> -butane, ethane and methane)	COFs (2,3-DhaTph and TpPa-1)	Classical DFT and GCMC simulations	Adsorption, selectivity, isotherms	Structural analysis	116
Acid gas removal from natural gas	10 top-performing COFs out of 70 000 COFs	Integrated molecular simulations with an ML approach	Deacidification/ Separation efficiency, structure–property correlation	Structural analysis	117
Adsorption behaviour of COF towards CO ₂	Polyarylether-based COFs (functionalized with –CN, –C(NH ₂) NOH and –COOH groups)	DFT and GCMC simulation, interaction energy and radial distribution analysis	Adsorption behaviour, gas storage and carbon capture, interaction energy, gas distribution analysis	Structural analysis	119
Separation of CH ₄ and C ₂ H ₆	2-dimensional imine COFs (1171 COFs with various configurations)	GCMC simulation	Adsorption and separation efficiency, structural parameters, nitrogen content, node type and monomer structure effect	Structural analysis	120
Separation of helium from natural gas	3D-Sp-COF-1	GCMC simulations and ML, MD simulations	Separation efficiency of binary mixtures of He/N ₂ and He/CH ₄ , structure–property correlation	Structural and dynamic analysis	113
Separation of CH ₄ and CO ₂	COF-based membrane	MD and GCMC simulations, thermodynamic analysis	Separation efficiency, effects of pressure, temperature and structure	Structural and dynamic analysis	122
Gas diffusion and adsorption behaviour of COF membranes (2D N-COF and 3D COF-300 membranes) for CH ₄ and CO ₂ ; CO ₂ /H ₂ separation performance	–NH ₂ group incorporated COFs	GCMC simulations, modelling	Adsorption behaviour, separation efficiency, experimental validation, membrane stability	Structural analysis	124
Adsorption of various gases like methane, hydrogen and argon	COF-102, COF-103, COF-105, and COF-108	GCMC simulations	Adsorption behaviour, experimental validation	Structural analysis	126
Hydrogen storage/separation of COF	COFs (boron–phosphorus and borophosphonate based)	GCMC simulations and DFT	Adsorption, storage and separation efficiency, structure–property correlation	Structural analysis	130,131
CH ₄ /H ₂ separation application	597 synthesized and 7737 hypo COFs	GCMC simulations, ML-assisted screening approach	Adsorption, separation efficiency, structure–property correlation	Structural analysis	132
COF incorporated Pebax polymeric matrix For separation of CO ₂ and CH ₄	Tentacle-like COF	DFT calculation and GCMC simulation	Separation efficiency, effect of pressure, temperature and COF loading on the membrane's efficiency, CO ₂ capture	Structural analysis	134
Controlled CO ₂ capture application	Photo-responsive COF-based architecture (NUT-105@COF-OCH ₃)	GCMC simulations	Adsorption capacity	Structural analysis	135
Hydrogen storage applicability of COFs	2902 COFs functionalized with Mg-alkoxide	Force-Field modelling, GCMC simulations, ML approaches	Hydrogen storage/adsorption capacity, binding energy, structure–property correlation	Structural analysis	136
CO ₂ separation	Hydrazone-based COF	DFT and MD approaches	Adsorption/separation efficiency, structure–property correlation	Structural and dynamic analysis	138
CO ₂ /N ₂ separation efficiency and gas adsorption performances of COF	Diamine-linked 2D COFs (building units: 1,3,5-tris(chloromethyl)benzene; linkers: <i>p</i> -diaminobenzene and hydrazine)	DFT and MD simulations	Adsorption capacity, separation efficiency, adsorption isotherms, permeability and selectivity	Structural and dynamic analysis	140



computational approaches for COFs or COF-based materials, investigating gas separation and storage with structural and dynamic analyses.

The adsorption study of gas molecules on COF can proceed in two phases. First, the COF or COF-based material structure can be validated through modelling, ensuring its structural correspondence to reality. Second, the adsorption simulation study can be used to investigate the gas uptake capability or efficiency of the structure. This approach correlates the material's structure with their properties, aiding efficient COF or COF-based material discovery, with superior gas storage and adsorption/separation ability.¹¹¹ Dong *et al.* demonstrated the role of BET and PXRD analysis for efficient monomer designs during the development of effective COF or COF-based materials.¹¹² Three types of two-dimensional isorecticular COFs such as 1,3,5-tris(4-aminophenyl) benzene (P-COF), tris(4-aminotriphenyl) amine (N-COF) and 2,4,6-tris(4-aminophenyl)-s-triazine (T-COF) were prepared from 1,3,5-benzenetricarbaldehyde (BTCA). The study compared the experimental PXRD (powder X-ray diffraction) pattern of COF structures with the pattern of structural simulations, confirming the material structure. The PXRD peaks for N-COF were observed at 6.8°, 11.8°, and 22.5°, while in the case of P-COF, the peaks were observed at 5.7°, 9.9°, 11.7°, and 25.1°. The experimental PXRD patterns match well with AB staggered and AA staggered stacking patterns of simulated N-COF and P-COF, respectively. Both structures possess low crystallinity owing to the presence of flexible amine linkages. Considering this fact, to improve the crystallinity of the COF structure, a rigid triazine-based linkage was introduced to produce T-COF. T-COF showed sharper PXRD peaks at 5.8°, 10.0°, 11.6°, 15.3°, and 25.9°, confirming better crystallinity with strong π - π stacking. Again, the Brunauer-Emmett-Teller (BET) analysis revealed the microporous structures of all three COFs, with pore volumes of 0.395, 0.442, and 0.549 cm³ g⁻¹, and BET surface areas of 692, 435 and 1149 m² g⁻¹. Nonlocal density functional theory-based calculation of pore width distribution showcased the framework's good match with the proposed crystalline structure. It was observed that the COF with planar amine linkages possess more crystallinity and high porosity, thereby exhibiting more hydrocarbon uptake capability. On the other hand, the N-COF possessing the smallest pore sizes demonstrates C₂H₆/CH₄ separation with the highest efficiency (as compared to the other two COF structures). Again, in another study, the gas storage and separation capability (for the mixture CO₂/CH₄) of N-COF, P-COF and T-COF was assessed employing GCMC and MD simulations.¹¹³ The adsorption isotherms (for C₂H₆ and CH₄, at 298 K) derived from the simulations are in good agreement with the experimental results, as reported by Dong *et al.*, for all three types of COFs. Owing to the smallest pore channels, N-COF demonstrated superior CO₂/CH₄ separation. Moreover, N-COF can effectively store CO₂ at low pressures, while at high pressures, P-COF and T-COF are the better candidates for CO₂ storage. This may be attributed to the dominant role of adsorbate-adsorbent interactions at low pressure and the key role of pore sizes in the case of high pressure. This type of experimental and

computational study may guide future studies on gas storage and separation, using various COFs or COF-based tailored materials.

A study investigates the interaction of N₂ and CO₂ gas with nitrogen-containing COFs by calculating the binding energies of the gas molecules with the COF architecture, employing the DFT approach.¹¹⁴ The capability of 580 synthesized and 891 hypoCOFs for the separation of carbon dioxide and hydrogen sulfide from natural gas and their selectivity towards them was explored utilizing computational investigations such as DFT and MD simulations (considering industrial-related environments).¹³ The study concluded that some of the synthesized and hypoCOFs exhibit excellent carbon dioxide and hydrogen sulfide selectivities outperforming conventional adsorbents. COFs hold emerging opportunities for their diverse applications in methane separation from industrially relevant gas mixtures including nitrogen, hydrogen and ethane, which were assessed through DFT calculations, MD simulations and GC simulations.¹¹⁵ The potential of 572 COFs in the separation of binary gas mixtures such as methane/hydrogen, methane/nitrogen and ethane/methane was investigated. These COFs outperformed traditional adsorbents in separation performance. The COF-incorporated membrane showed excellent hydrogen permeability with favourable selectivity, which makes them superior to polymeric and MOF-based membranes. The adsorption of gases (*n*-butane, ethane and methane) in COFs (2,3-DhaTph and TpPa-1) was modelled using classical DFT and GCMC simulations. The findings showed good agreement between the two methods, when studied under the condition of pressure up to 50 bars. The study underscores the significance of classical DFT for the prediction of adsorption of long hydrocarbons in the COF architecture.¹¹⁶ Integrating molecular simulations with an ML approach was followed for the rapid screening of 10 top-performing COFs out of 70 000 COFs, for the application of deacidification (acid gas removal from natural gas).¹¹⁷ The study showed the high separation performance of COF-incorporated membranes surpassing the efficiency of the MOF fillers. In another GCMC simulation study, a novel MOF was designed where carbon flakes were used to modify IRMOF-1, which showed enhanced gas storage efficiency.¹¹⁸ The circular carbon flakes demonstrated the best CO₂ storage capacity, while rhombus flakes exhibited the best H₂ storage. These findings could greatly benefit the design of novel, highly efficient COFs/MOFs for gas storage/separation applications. DFT and GCMC simulations were employed to evaluate the adsorption behaviour of polyarylether-based COFs (functionalized with -CN, -C(NH₂) NOH and -COOH groups) towards CO₂, thereby showcasing their promising potential in gas storage and carbon capture.¹¹⁹ Using GCMC simulation, an investigative study was carried out to analyze the potential of the 2-dimensional imine COFs (1171 COFs with various configurations) for the separation of methane and ethane.¹²⁰ MD simulations and ML were employed in a recently reported work to screen out the best-performing COF or MOF-based membranes for the separation of helium from natural gas. Among all, 3D-Sp-COF-1 exhibited excellent helium adsorption ability



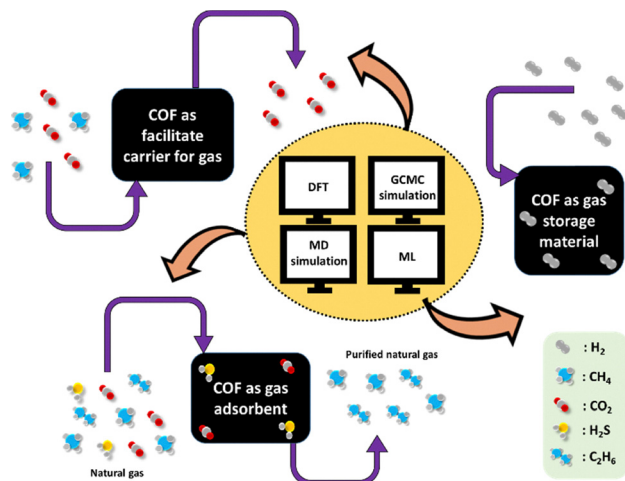


Fig. 5 Representation of the most commonly used methods for various gas storage and separation applications.

and separation efficiency for the binary mixtures of He/N₂ and He/CH₄.¹¹⁰ The findings emphasized that the COFs having 3D structures, high surface area and larger pore volumes play a major role in effective helium separation. The role of computational simulations and ML approaches in exploring the effectiveness of COFs/MOFs and further design strategies of such materials for gas separation, storage and delivery applications was reported.¹²¹ Fig. 5 represents the most commonly employed computational approaches to investigate the gas storage and separation performance of COFs.

Strategies for the separation of CH₄ and CO₂ using a COF-based membrane were discussed employing MD and GCMC simulations, investigating the potential impact of temperatures, pore sizes and functional groups in enhancing the separation process.¹²² Gas diffusion and adsorption behaviour within -NH₂ group-incorporated COF architectures (for CH₄ and CO₂) and their interactions were evaluated.¹²³ Upon incorporation of -NH₂ group into the COF architecture, CO₂ adsorption capability improved; however, the dynamic behaviours of the gas molecules within the architecture remained unchanged. The GCMC simulation further validates the excellent CO₂-H₂ separation performance of a COF-based self-standing membrane possessing enhanced H₂ permeance.¹²⁴ In another study by Keskin *et al.*, it was observed that COF-6 outperformed various nanoporous materials, towards the effective separation of CH₄ and H₂.¹²⁵ The adsorption and diffusion phenomena of gas molecules within various COF frameworks were analyzed employing GCMC and equilibrium molecular dynamics (EMD) simulations. In another study, by considering COF-102, COF-103, COF-105, and COF-108 architectures, the adsorption of various gases such as methane, hydrogen and argon was presented.¹²¹ Out of the COFs used, COF-102 performed with exceptional methane storage capability, whereas most of the COFs exhibited strong hydrogen adsorption behaviour. The methane adsorption behaviour of various 2D and 3D COFs, under different pressure conditions, with their insight mechanisms was discussed employing an experimental study

validated by computer simulations.^{126,127} The highest methane storage capacity by COF-1 (2D) and methane delivery efficiency by COF-102 (3D) and COF-103 (3D) were showcased in the study.

Key factors such as pore size optimization, impregnation and structure modifications responsible for hydrogen storage applications of porous materials (including MOFs and COFs, both 2D and 3D), and their improvement strategies were explored using quantum calculations, MD simulations and GCMC simulations.^{128,129} The efficient hydrogen storage/separation abilities of novel COFs (boron-phosphorus and borophosphonate-based) possessing high valency were investigated by harnessing DFT and molecular simulations, under different pressure and temperature conditions.^{130,131} An ML-assisted screening approach was carried out computationally to assess the potential of 597 synthesized and 7737 hypoCOFs (*i.e.* screened out from 69 840 hypothetical COFs), in CH₄/H₂ separation applications.¹³² Identifying the best-performing COFs (considering adsorption and regeneration capability), the impact of structural features and pressure variations on the adsorption capability of the COFs was emphasized in the study, employing GCMC simulations. The findings demonstrated better CH₄/H₂ separation efficiency of the hypothetical COFs than that of the synthesized COFs and MOFs. The quick separation performance predictions based on the structural features of the COFs were carried out using ML models. These studies suggest various pathways and strategies for accelerating further advancements of COFs in hydrogen separation and storage applications.

The role of fillers including COFs and MOFs in polymeric mixed matrix membranes was reported for enhanced separation of gases with the help of high-throughput computational screening (to find out the best filler materials), where the interactions of the gas molecules with the membrane were evaluated using molecular simulations.¹³³ Tentacle-like COFs were incorporated as fillers into the Pebax polymeric matrix (used as a mixed-matrix membrane) to obtain improved separation of CO₂ and CH₄.¹³⁴ The COFs introduced various nano-traps for capturing CO₂, with the best CO₂ separation performance observed at 3% COF loading into the polymeric matrix. The significance of COF loading into the polymeric matrix was demonstrated by the enhanced separation efficiency of the COF-incorporated membrane (up to 73.4% and 32.9%) as compared to the pure polymeric membrane, which was further validated by DFT calculations and GCMC simulations.

A photo-responsive COF-based architecture (NUT-105@COF-OCH₃) was designed by Yang *et al.* for improved and controlled CO₂ capture application (achieving up to 29.2% of CO₂ adsorption, with better light response).¹³⁵ The alternation of pore sizes upon irradiation of UV- and vis-light (leading to regulated CO₂ adsorption) was further illustrated using GCMC simulations. The hydrogen storage applicability of COFs functionalized with Mg-alkoxide was explored in another study using thorough computational screening employing simulations and ML approaches.¹³⁶ Enhanced CO₂ uptake capacities of the COFs, concerning the temperature and pressure variations (and the



impact of functionalization), were predicted, showing that COFs having small pore sizes exhibited better CO₂ capture capabilities.^{132,137}

The role of hydrazone-based COFs in CO₂ separation as well as capture was analyzed using DFT and MD approaches. Their tunable structural properties, strong CO₂ interaction and effective sieving effect position them as potential candidates in CO₂ separation and storage applications.¹³⁸ With the advantages of illustrating the time-dependent dynamic nature of gas molecules within the COF framework, MD simulations can complement the GCMC simulations.¹³⁹ The MD approach enables the analysis of gas transport mechanisms within the framework, including the diffusion of gas molecules and the influence of the structural framework on these processes.

Employing DFT and MD simulations, the CO₂/N₂ separation efficiency and gas adsorption performances of diamine-linked 2D COFs (building units: 1,3,5-tris(chloromethyl)benzene; linkers: *p*-diaminobenzene and hydrazine) were discussed in detail. By maintaining the typical thermodynamic conditions of industrial flue gas after combustion, the remarkable CO₂

capture ability of the COFs was assessed.¹⁴⁰ In order to identify and optimize the crucial parameters (such as pores and surface areas), for efficient hydrogen storage applications, a combined approach of *ab initio* calculations, MD and GCMC simulations was harnessed.¹⁰⁶

These investigations may pave the way for future advancements of novel COFs or COF-based materials endowed with high hydrogen storage capabilities, for cutting-edge research. The impact of tiny porous architectures of COFs in enhancing their CO₂ separation and capture ability was also demonstrated, utilizing triazine-based benz-bis(imidazole)-bridged COFs.¹⁴¹ Again, in another work, ML-accelerated high-throughput screening was carried out, figuring out the top-performing COFs and showcasing their CO₂ separation potential from flue gases. The study revealed the role of various linkages and pore sizes/distributions in COF architectures, influencing CO₂ capture capabilities. The significance of 2D COFs possessing fluoro groups and imide linkages (remarkably enhancing their selectivity towards CO₂) was reported in the study, and it was also observed that 3D COFs with



Fig. 6 Adsorption of CO₂ and N₂ on IL-COF-1 and COF-366, showing (a) only CO₂ and (b) gas mixture. Reproduced from ref. 143 with permission from the Royal Society of Chemistry, Copyright 2014.



approximately 1 nm pore diameter exhibited excellent CO₂ capture ability.⁹² Modelling adsorption of volatile organic compounds (VOCs) on COFs was carried out utilizing DFT calculations and MD simulations, revealing the potential of COFs in the mitigation of indoor air quality issues.¹⁴²

The studied VOCs in the work include benzene, toluene and formaldehyde, whose selectivity and adsorption on 37 screened-out COFs (obtained from experimental data) were reported (over N₂), providing insights into adsorption mechanisms. The adsorption of CO₂ and N₂ on IL-COF-1 and COF-366 was discussed,¹⁴³ emphasizing the importance of COFs in gas separation applications, as shown in Fig. 6. These studies may pave the way for further advancements of COFs in broad-scale gas storage/separation, and hydrocarbon separation/purification applications.

MD simulations were further employed to analyze the removal of CCl₄ from gas mixtures/air effectively, using COFs, ZIFs and IRMOFs. The effective role of the architectural design of the materials in enhancing adsorption-based separation was also discussed.¹⁴⁴ Additionally, ML integrated with simulation approaches was employed to study the industrially challenging separation of ethane from ethene using optimal COFs.^{145,146}

Further, the study emphasized the substantial impact of COF density on the selectivity of the gases and enhanced selectivity of COF-303 upon introduction of fluorine.¹⁴⁵ Again, optimal fluorinated ionic liquid (FIL)-COFs were screened, using ML approaches, for effective separation of ethane and ethene.¹⁴⁶ In another study, employing the XGB model of ML, four COFs were identified from the CURATED database for the effective separation of SF₆ and N₂. Subsequently, employing GCMC simulations and DFT calculations, the adsorption

performance of the COFs and underlying mechanisms were explored.⁹³ A combined approach of ML and molecular simulations was employed in another study to assess the gas adsorption properties of 69 784 COFs, from various gas mixtures such as O₂/N₂, CO₂/N₂, CH₄/N₂, CH₄/H₂, CO₂/H₂ and CO₂/CH₄, revealing the chemical and structural features of the top-performing COFs (Fig. 7).^{18,147}

These studies collectively confirmed/highlighted the immense potential of various COFs in gas separation/storage and carbon capture applications, guiding future advancements in COF design.

3.2. Liquid separation and purification

The MD approach was utilized to evaluate the water desalination application of a three-dimensional COF framework, named 3D-OH-COF, emphasizing the underlying mechanism of the separation process.¹⁴⁸ In this study, the COF demonstrated excellent salt rejection ability (up to 100%) with very high water flux, even better than commercial RO membranes. The transport of ions as well as water through the nanochannel of COF was also examined employing MD simulations, highlighting their significant potential in desalination applications.¹⁴⁹ In another study, Guan *et al.* discussed the effect of layer spacing on the ion's interaction and water flow during desalination using COF-based membranes.¹⁵⁰ The effective removal of mercury (up to 99%) from acidic water was obtained when a COF-supported silver nanoparticle was employed as an adsorbent material (further validated by DFT calculations), which was highly stable and regenerable in nature.¹⁵¹

By conducting DFT calculations, the boroxine-based COFs were tested for their use in the removal of toxic heavy metals



Fig. 7 Selectivities of the COF materials for CO₂/CH₄, CO₂/H₂, CO₂/N₂, CH₄/H₂, CH₄/N₂, and O₂/N₂ separations. Selectivities of 68 724 hypoCOFs (left panel) and 1060 CoRE COFs (right panel) were computed at (a) 0.1, (b) 1, (c) 5, and (d) 10 bars. Reproduced from the ref. 18. Copyright © 2025, American Chemical Society.



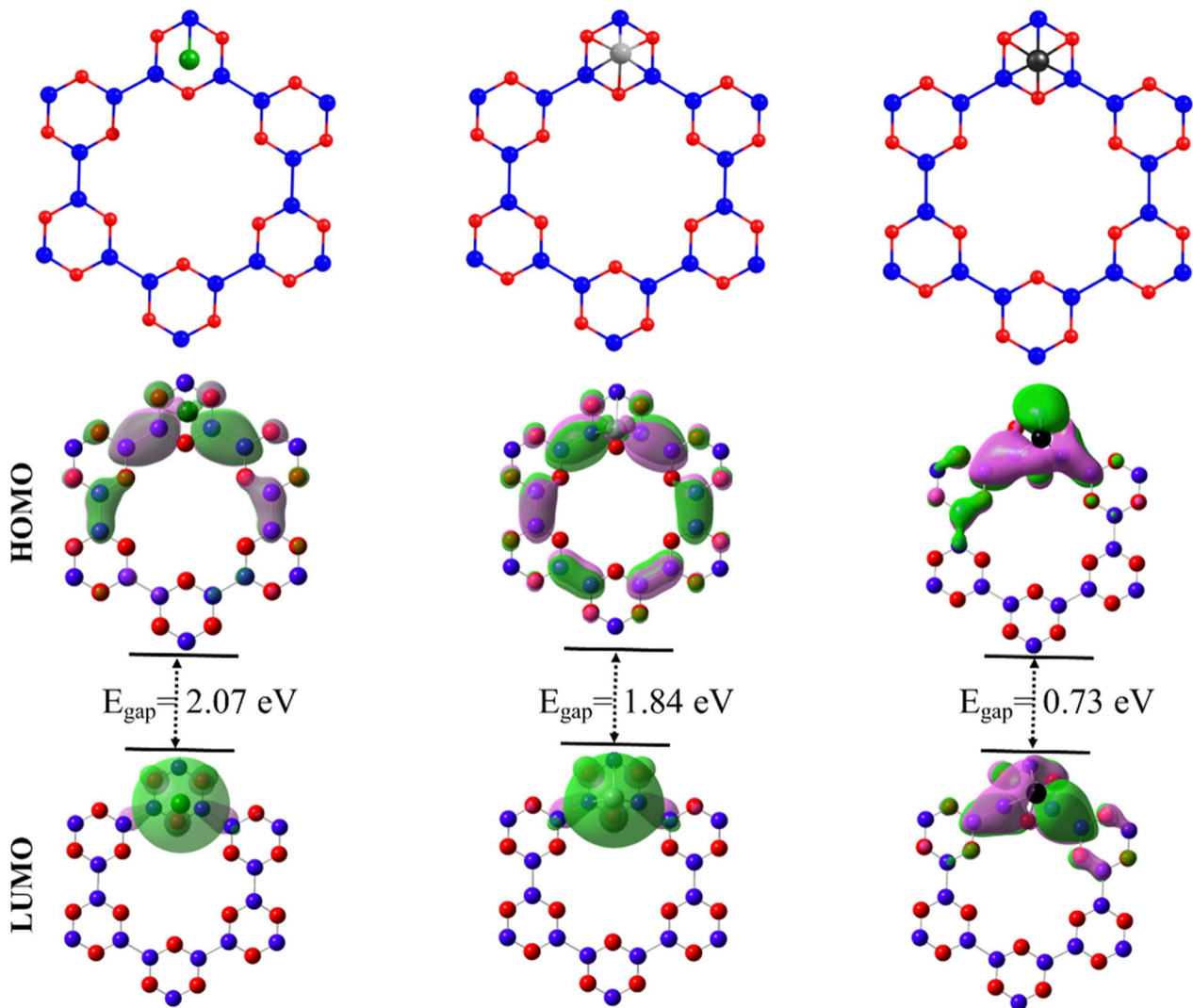


Fig. 8 Optimized structures, HOMOs and LUMOs, and E_{gap} values of the heavy metal@boroxine complexes. B, O, Cd, Hg, and Pb atoms are shown in blue, red, green, gray, and black colors, respectively. Reproduced from ref. 152 with permission from Elsevier, Copyright 2021.

(Hg(II), Pb(II) and Cd(II)) from wastewater, and the charge transfer from boroxine to metal ions was discussed (as shown in Fig. 8).¹⁵² It was observed that upon adsorption of the metal ions, the HOMO–LUMO gap reduced, enhancing the electron transport with effective binding affinity between the COF and the heavy metal ions. Again, the utilization of various COFs and COF-based materials for the removal of palladium,¹⁵³ lead²⁹ and Cr(VI)¹⁵⁴ from water streams and the recovery of Au(I) from the solution of thiosulfate¹⁵⁵ were reported using experimental and DFT approaches. These studies underscore the remarkable versatility of COFs in environmental remediation and recovery of precious metals, providing an in-depth understanding of the separation mechanisms.

The dye (methylene blue) adsorption properties of Schiff base network1 (SNW1) (a series of COFs synthesized by Mullen *et al.*¹⁵⁶ via the reaction of terephthalaldehyde and melamine) were evaluated using DFT, which showed a thermodynamically favourable spontaneous and exothermic interaction of the

COFs with the dye molecules.¹⁵⁷ Zhao *et al.* reported a pH-responsive COF-based membrane (fabricated through an interfacial polymerization approach) that shows colour change with pH, for effective removal of dyes and salts from the wastewater stream. The membrane showed good removal efficacy with excellent water flux, especially at high pH values. The MD simulation was carried out to demonstrate the separation of dye molecules using the COF membrane architecture.¹⁵⁸ The effective role of COF-1 and COF-2 nanostructures in the removal of alizarin dye and its derivatives from the aqueous phase was studied using molecular simulations. COF-1 exhibits better interaction with the dye molecules, resulting in more effective removal, where π – π and van der Waals interactions played a significant role.¹⁵⁹ The fabrication of TpPa COF membranes and their performance were evaluated for the separation of dyes from industrial wastewater.^{9,160} From the observation, it was concluded that π – π interaction and surface adsorption are the key factors behind the enhanced separation



performance of the membrane. The separation efficiency of the membrane with respect to the structure of dye molecules and the membrane thickness was also emphasized in the study.⁹ The impact of functional groups and pore sizes on the dye (bisphenol A) adsorption efficiency of COFs (in both liquid and gas environments) was discussed by Wang *et al.* using DFT, GCMC and MD simulations.¹⁶¹ The COFs functionalized with -COOH groups stand best in terms of the dye uptake capacity, and it was seen that high temperatures reduce the adsorption performance. The removal of dye pollutants (such as Congo red, gentian violet cat, brilliant green, methyl orange, rhodamine B, tonium chloride and methylene blue) from water using 1,3,6,8-tetrakis(4-formylphenyl)pyrene-COF (3D) was evaluated using computer simulations. DFT and MD simulations

(with experimental study) were carried out to study the adsorption of bisphenol-A¹⁶² and the removal of malachite green from the water stream,¹⁶³ using COF-based materials. Harnessing MD and well-tempered metadynamics simulations, the dye removal capability of 3D TFPPy-COFs (with underlying dye adsorption mechanisms) was analyzed for different dye contaminants including Congo red (CR), rhodamine B (RhB), brilliant green (BG), gentian violet (GV), tonium chloride (TC), methylene blue (MB) and methyl orange (MO) from the aqueous phase (as shown in Fig. 9).¹⁶⁴ From the metadynamics simulations negative free energy surface (FES) was observed, indicating energetically favourable interactions, as the proposed dye molecules approach the COF architecture. The COFs demonstrated rapid adsorption and promising



Fig. 9 (i) Final snapshots of the dyes/COFs (water molecules are not shown for clarity). (ii) Interaction energy between dye molecules and COFs (total, Lennard-Jones (L-J), and Coulombic energies for the adsorption). (iii) Initial snapshot of the studied systems for COFs/dyes. Color code-carbon: cyan, hydrogen: gray, sulfur: yellow, and nitrogen: blue. (iv) FES patterns for transferring dye molecules (a–e: MO, GV, BG, TC and RhB) towards the COFs materials. Reproduced from ref. 164 with permission from Springer Nature, Copyright 2024.



the COF), in improving the drug adsorption efficacy and enhancing drug recovery.¹⁸⁶ Molecular dynamics and metadynamics simulations were carried out to monitor the impact of -OH functionalization on the 2D COFs during the adsorption of doxorubicin, with pi-pi interactions and hydrogen bonding interactions playing a critical role. In this case, the drug release also favoured an acidic environment, reinforcing the therapeutic applications of the COFs.¹⁸⁷

Furthermore, a study investigated the therapeutic potential of 2D COFs in treating Alzheimer's disease through a multi-scale simulation approach.¹⁰⁷ Alzheimer's disease is mainly caused by the aggregation of β -amyloid ($A\beta$) protein. To

mitigate this, a multi-scale computational strategy was adopted to assess the interaction of the 2D COF with the $A\beta$ protein, thereby disrupting their aggregation. Apparently, surface area and functional groups play a crucial role in the inhibitory impact of the 2D COFs. The significance of COF-based architecture in photodynamic therapy applications (having exceptional antimicrobial activities) was studied through experimental investigation and MD simulations.¹⁸⁸ A novel amine-based COF was reported for its excellent drug loading capacity (31.32%) and carboplatin (antineoplastic drug) release efficiency at pH 5, making it effective in anticancer therapy. The adsorption mechanism of the drug on the COF architecture,



Fig. 10 Schematic showing the potential of COFs in various drug delivery applications.



COF S-scheme heterojunctions,²¹¹ protonated Z-scheme CdS-COF heterojunction,²¹² COF/2D g-C₃N₄ heterostructure,²¹³ arylboron functional COFs,²¹⁴ Pt single atom²¹⁵ and Pt cluster-incorporated COFs²¹⁶ and isoelectronic organic dye-based COFs²¹⁷ for photocatalytic H₂ production was extensively discussed. These study offers an in-depth understanding of COFs in photocatalytic H₂ production applications, providing insights into the charge transfer pathways/mechanisms.^{211–217} Mourino *et al.* reported a screening approach (based on low-cost DFT calculations) on the CURATED COFs database and identified the most promising COF candidates (13 numbers) having effective water-splitting capability. The study also highlighted the best-performing COF materials (N_x-COF (x = 0–3)) for potential applications in H₂ evolution.²⁷ The impact of multipolar structures, functional groups and hydrazone linkage density on photocatalytic hydrogen peroxide evolution was also discussed using experimental and DFT-based approaches (Fig. 11).^{218–220}

Electrocatalytic nitrate reduction and synthesis of ammonia by copper-bipyridine 2D COF films were investigated using experimental and theoretical approaches, where it was found that the structural features of the film effectively enhanced mass transfer and ammonia production.²²¹ Computer simulations demonstrated the photocatalytic conversion of nitrogen into ammonia using 3D COFs. After screening the experimental COF database, COF-300 is identified as the most promising candidate owing to its electronic properties and nitrogen uptake capabilities.²²² In an attempt to enhance the (aerobic) photocatalytic oxidation processes, COFs with three-motif molecular junctions were designed and studied employing experimental study and DFT calculations.²²³

It was observed that the best-performing COF effectively converted benzylamine into imines within 2.5 hours with 99.9% conversion efficiency, owing to high light absorption capability and a synergistic electron transfer pathway. Excellent bisphenol degradation was achieved using the photocatalytic

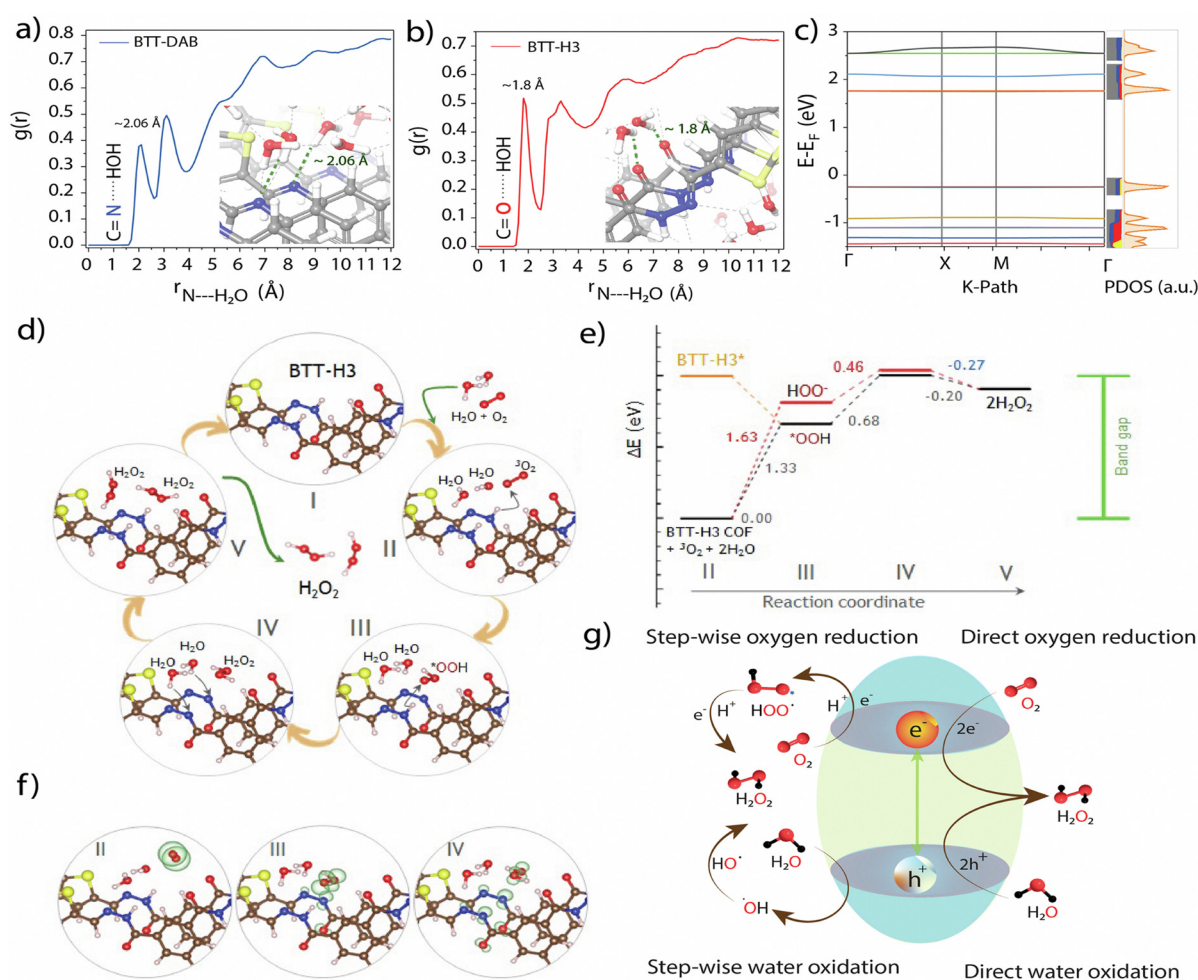


Fig. 11 Photocatalytic H₂O₂ evolution study employing computational analysis: (a) N center of BTT-DAB (Benzo[1,2-*b*:3,4-*b'*:5,6-*b''*]) trithiophene-2,5,8-tricarbaldehyde and 4-diaminobenzene) COFs, (b) O center of BTT-H3 (hydrazone-linked COF), (c) band structure and projected density of states (PDOS) for BTT-H3, (d) molecular mechanism of the generation of H₂O₂ from oxygen and water at the hydrazone center of the BTT-H3 COFs, (e) electronic energy profile of the hydrogen peroxide generation reaction path, (f) spin densities (semi-transparent light green blobs) of the radical species, and (g) representation of the overall photocatalytic mechanism of H₂O₂ generation through the water oxidation reaction and oxygen reduction reaction. Reproduced from ref. 220 with permission from Springer Nature, Copyright 2025.



activity of COFs utilizing an integrated approach of experimental studies and DFT calculations.²²⁴

Recent studies have focused on the development of COFs and metal-based COFs for numerous applications, including environmental remediation, reduction of CO₂ and extraction of uranium from seawater, which have been extensively evaluated using experimental, DFT and ML approaches.^{225–231} These studies may play a crucial role in leveraging advanced design strategies for next-generation COF-based photocatalysts and

their future advancements in the domains of optoelectronic, photocatalytic, and environmental remediation applications.

3.5. Photovoltaics and sensors

The electronic properties and charge dynamics of 2D aza-COFs were analyzed with the help of time domain DFT calculations and non-adiabatic MD simulations. The direct band gap, prolonged electron-hole recombination, and exceptional light absorption of the COF and its potential for effective



Fig. 12 (i) Optimized structures of the GCS with the pure and functionalized COFs, (ii) optimized structures of the stable configurations of the stable complexes (complexes of the COFs with GCS) studied at the M06-2X/ LanL2DZ level of theory, and (iii) 3D NCI isosurfaces and 2D RDG scatter maps for all the studied complexes. Reproduced from ref. 233 with permission from Elsevier, Copyright 2025.



solar-energy harvesting and future applications in photovoltaic devices were also discussed in the study.¹⁷⁸ A novel subphthalocyanine-based 3D COF, co-crystallized with fullerene, displayed enhanced photovoltaic performance, achieving a balance between long-range order and surface area.²³² The evaluation of electronic properties, insights into charge transfer, structural optimization and separation mechanisms of the COFs were explored using DFT and MD simulations. The recent advancements in photovoltaic applications utilizing COFs and MOFs were also reviewed in another study, owing to their exceptional stability and tunable electronic properties.²³³

Furthermore, a DFT study explored the detection of nitroaniline isomers (which are toxic in nature), using 2D B₆N₆ (boron nitride) COFs.²³⁴ The study revealed the binding behaviour of the COFs with the nitroaniline isomers through weak van der Waals interaction, which pointed out their potential towards sensor applications. The chemical and thermal properties of imine-linked COFs were characterized through computational and experimental approaches.²³⁵ B₆N₆ COFs were reported for their effective sensing capabilities in detecting cathinone drugs (showing significant conductivity changes), and adsorption energies were calculated using the DFT approach.²³⁶ Various COF-based architectures were reported employing experimental study, DFT calculations, GCMC and MD simulations for their use in sensor applications. These include selective sensing of nitrogen dioxide,²³⁷ ethane and carbon monoxide,²³⁸ dopamine in serum,²³⁹ HCl vapours,²⁴⁰ formic acid,²⁴¹ nitric oxide and hydrogen sulfide,²⁴² and volatile organic compounds.²⁴³ Employing DFT calculations (utilizing the M06-2X/Gen/6-311G(d)/LanL2DZ method), the adsorptive behaviour of glyphosate (GCS), one of the most commonly used herbicide, on the TpBD-based COF architecture and its potential sensing application were studied, as shown in Fig. 12.²⁴⁴ The study explores the impact of various functional derivatives such as -NO₂, -NH₂, -CF₃ and -COOH, when incorporated into the COF, in the adsorption process. Out of all the studied systems, GCS@COF-NH₂ possessed the highest negative value of adsorption energy. Two-dimensional reduced density gradient (RDG) scatter plots and 3D non-covalent interaction (NCI) isosurface plots were visualized, revealing the variations of interaction in all the studied complex systems. In 3D visualization as represented in Fig. 12, the green and blue regions indicate weak vdW and H-bonding interactions, respectively, whereas the repulsive forces in the systems are indicated by the red patches.

4. Conclusion

COFs are emerging as a groundbreaking class of materials in the current scientific research, showcasing their significant potential in various applications such as gas separation/storage, wastewater treatment, drug delivery, photocatalysis, sensing, photovoltaics and environmental remediation. To advance the understanding of COFs, computational chemistry plays an imperative role, providing molecular-level insights and highlighting the promising features of COFs that make them suitable for a broad range of research domains. Computational

chemistry is a powerful tool providing an in-depth evaluation of interaction/adsorption mechanisms, electronic properties and performance–structure relationships. The impact of the pore sizes, topology, functional groups and configurational modifications of COFs on the gas storage, adsorption and separation processes and catalytic activities, from the perspective of computational chemistry, was discussed in this review article.

Future studies should focus on linkage engineering to enhance the stability and properties of COFs, making them more effective in real-world applications. Proper planning of a functionalization strategy with precise positioning within the COF architecture is indeed valuable without disturbing its original framework, which may be carried out through host–guest interactions, metalation, and click chemistry. Further research should be carried out on chemical locking (by converting the linkages to a more stable form) and protective coating of COFs, which may enhance their lifespan to a great extent, even under harsh physical and chemical conditions. The designing of hybrid materials should also be anticipated, by integrating COFs with other materials such as graphene, CNT and MOFs, resulting in advanced composites with unprecedented properties and performance. Upcoming studies should aim to develop computational high-throughput screening approaches to find out the most suitable COFs for particular applications. Moreover, advanced 3D printing techniques could be employed to build COFs with complex designs and tailored properties for specific applications. Machine learning integrated with computational modelling can facilitate the discovery of novel COF-based materials by predicting their behaviour and properties. Overall, future research should focus on computational and experimental strategies for tuning pore sizes, chemical functionalities and surface area of COFs or COF-based materials, to achieve their large-scale application in industrial as well as scientific domains.^{16,245}

It is noteworthy that, in contrast to several advancements, hurdles remain in combining computational and experimental findings and attaining effective/accurate multiscale modelling, which should be properly addressed to fully unveil or unlock the exciting potential of COFs in diverse domains. The potential of various advanced computational strategies was critically reviewed while addressing the optimization approaches and key challenges during the COF design, ultimately guiding the design and development of tailored COF-based materials for numerous applications. Bridging ML and simulations with experimental feedback may lead to an era of more rational COF design. Meanwhile, we believe that this study can serve as a stepping stone for future advancements, accelerating the design of novel next-generation COF-based materials (both experimentally and computationally) and paving the way for their applications in real-world scenarios, tackling global challenges.

Author contributions

Parashmoni Rajguru: drafting the original manuscript, editing, reviewing, formal analysis, software. Prarthana Bora: software,



formal analysis. Chinmoy Bhuyan: software, formal analysis. Swapnali Hazarika: conceptualization, investigation, writing – review & editing, supervision.

Conflicts of interest

The authors declare that this work has not been published previously, and it is not under consideration for publication elsewhere. The authors also declare that they have no conflicts of interest.

Data availability

All the relevant data have been presented in the manuscript.

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