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Perspective

Covalent-organic frameworks: An intelligent platform for photocatalytic uranium reduction and separation

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ABSTRACT

Covalent-organic frameworks (COFs) have attracted growing interest as photocatalytic materials for uranium extraction because of their tunable structures and adjustable optoelectronic properties. This perspective summarizes recent progress in COF-based systems for the selective capture and photoreduction of U(VI), with a focus on molecular design strategies such as band-structure regulation, donor-acceptor framework construction, and interfacial functionalization to improve visible-light utilization and charge separation. Key mechanistic aspects of photoinduced uranium conversion, including electron-transfer pathways, uranium valence evolution, and interfacial reaction kinetics, are discussed. Current challenges, such as uncertainty in reduction products, incomplete mechanistic understanding, and limited cycling stability, are also addressed by highlighting the role of *in situ* characterization combined with theoretical analysis. Finally, the potential of artificial intelligence-assisted design and the development of macroscopic COF forms, such as aerogels and membranes, are briefly discussed as routes toward practical uranium extraction.

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1. Introduction

Nuclear power is a prospective energy resource with high-energy density and low-carbon emission properties [1]. Uranium is crucial to the sustainable development of nuclear energy [2]. Recently, uranyl capture from either nuclear wastewater or natural seawater has been studied extensively and mainly relies on the strong selective binding affinities/chelating to UO_2^{2+} using chelating polymers, porous inorganic or carbon-based materials, covalent-organic frameworks (COFs) and metal-organic frameworks (MOFs) [3–5]. However, hexavalent uranyl (U(VI)) has high toxicity and mobility. Reducing U(VI) to U(IV) (UO_2) precipitates can significantly enhance its complexation and separation capacity, obviously enhancing uranyl adsorption and separation efficiency. In recent years, techniques with low chelation strength and high catalytic reduction ability for U(VI) reduction to U(IV) have been investigated intensively, such as electrocatalysis [6], photocatalysis [7], adsorption-photocatalysis [2], and electrochemical technologies. Given the natural sunlight advantages, photocatalysis without sacrificial reagent has green, solvent free, highly efficient, and sustainable properties, shows promising potential in photocatalytic reduction of U(VI) to achieve high U(VI) extraction.

Owing to the complex environmental conditions in seawater and wastewater, photocatalytic induced uranyl reduction and extraction is highly challenging, especially because solution conditions and catalyst types significantly affect the final form of uranium products. In practical seawater systems, biofouling, interference from natural organic matter, and strong competition from carbonate species can further hinder uranyl adsorption and photoreduction processes. Uranyl ion has two axial oxygen atoms, with the equatorial plane typically accommodating 5 or 6 coordinated atoms or hydrated waters at bond distances ranging from 0.2 nm to 0.3 nm [8], making it with variable coordination types and valence states. This intrinsic characteristic of the uranyl ion leads to relatively complex photo-induced reaction pathways and product distributions. Deep understanding of the photocatalytic process and the characterization of photocatalytic induced uranyl products from seawater and wastewater are of great significance for highly selective extraction of U(VI) from complex systems.

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Traditional adsorbents (such as polymers, amorphous carbon) and photocatalysts (such as TiO_2) have drawbacks of low specific surface area, poor selectivity, slow kinetics, weak adsorption capacity, and limited selectivity for the adsorption-photoreduction of U(VI). In photocatalytic uranium extraction, the role of adsorption extends beyond simple preconcentration, as the adsorption kinetics of uranyl species determine their surface coverage and residence time at the catalytic interface, thereby regulating interfacial electron transfer and photoreduction efficiency [9,10]. According to our recent investigation, the high uranium extraction ability could decrease photocatalytic delay time and increase the reaction rate obviously [11]. Although adsorption influences the efficiency of photocatalytic uranium extraction, the photocatalytic reduction process is the decisive factor governing overall extraction efficiency. As a result, the development of efficient photocatalytic technologies for U(VI) removal continues to face significant challenges. Firstly, most photocatalysts suffer from limited light absorption, as visible and near-infrared light, which constitute the majority of solar energy, possess insufficient excitation capability. Secondly, inefficient generation and separation of electron-hole pairs further restrict photocatalytic performance due to the high binding energy and poor dissociation of singlet excitons, which often decay *via* fluorescence. Lastly, efficient conversion, transport, and utilization of excitons at reactive sites remain critical challenges.

The performance of U(VI) adsorption and photocatalytic extraction is commonly evaluated using metrics such as adsorption capacity, uptake kinetics, photoreduction efficiency, selectivity, and cycling stability. However, these parameters are strongly influenced by experimental conditions, and values obtained in simplified systems may overestimate performance in realistic seawater environments due to carbonate competition and organic matter interference. Therefore, a comprehensive assessment should consider multiple complementary metrics and their limitations when extrapolating laboratory results to practical applications.

2. Representative advances of COFs in photocatalytic extraction of U(VI)

COFs have emerged as a promising class of photocatalytic materials to address these intrinsic limitations. Owing to their modular and highly tunable molecular structures, COFs allow precise regulation of π-conjugation length, band structure, and functional groups, thereby enabling enhanced visible light absorption and more efficient utilization of the solar spectrum [12,13]. In addition, the periodic and ordered framework of COFs facilitates exciton migration and charge separation by providing well defined transport pathways, effectively suppressing electron-hole recombination [14,15]. The incorporation of donor-acceptor (D-A) building blocks and redox-active sites further promotes internal exciton conversion and directional charge transfer toward catalytic centers [16]. Moreover, the high specific surface area and uniformly distributed active sites of COFs are advantageous for the adsorption and subsequent photoreduction of U(VI), making them particularly suitable for photocatalytic uranium extraction under complex aqueous conditions. The timeline of COF-based photocatalytic uranium extraction is shown in Fig. 1. The application of COFs in uranium extraction has evolved from early adsorption-based systems relying on pore confinement and coordination interactions to functionalized and photoactive COFs with enhanced affinity and selectivity. More recently, rationally designed COFs integrating D-A architectures and redox-active sites have enabled efficient photocatalytic uranium capture and reduction in complex aqueous environments [17,18].

To improve the adsorption capacity of visible and near-infrared light by COFs, various strategies have been investigated. According to the literature recently, anchoring metal ions, modification functional groups, tuning D-A structures or introducing nitrogen-containing or oxygen-containing active sites to COFs may become effective methods. In our recent investigation, a new visible-light

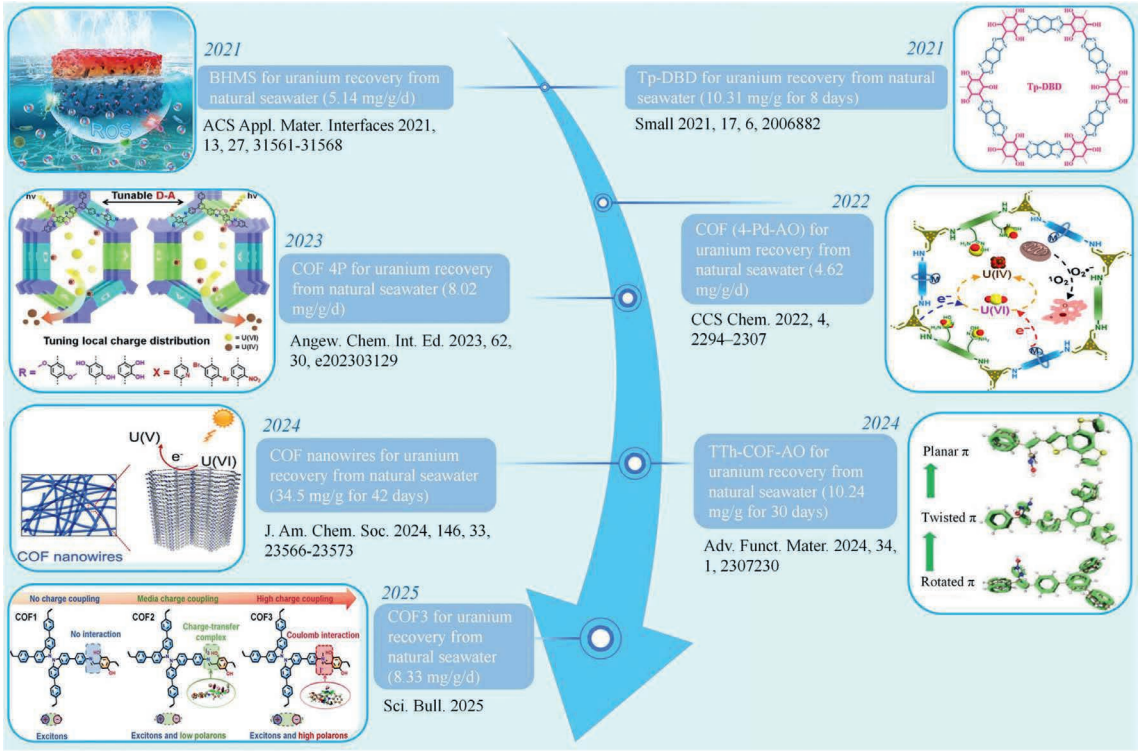


Fig. 1. Timeline of COFs applications in photocatalytic uranium extraction. Reproduced with permission [17]. Copyright 2021, American Chemical Society. Reproduced with permission [18]. Copyright 2021, Wiley. Reproduced with permission [26]. Copyright 2024, Wiley. Reproduced with permission [32]. Copyright 2024, American Chemical Society.

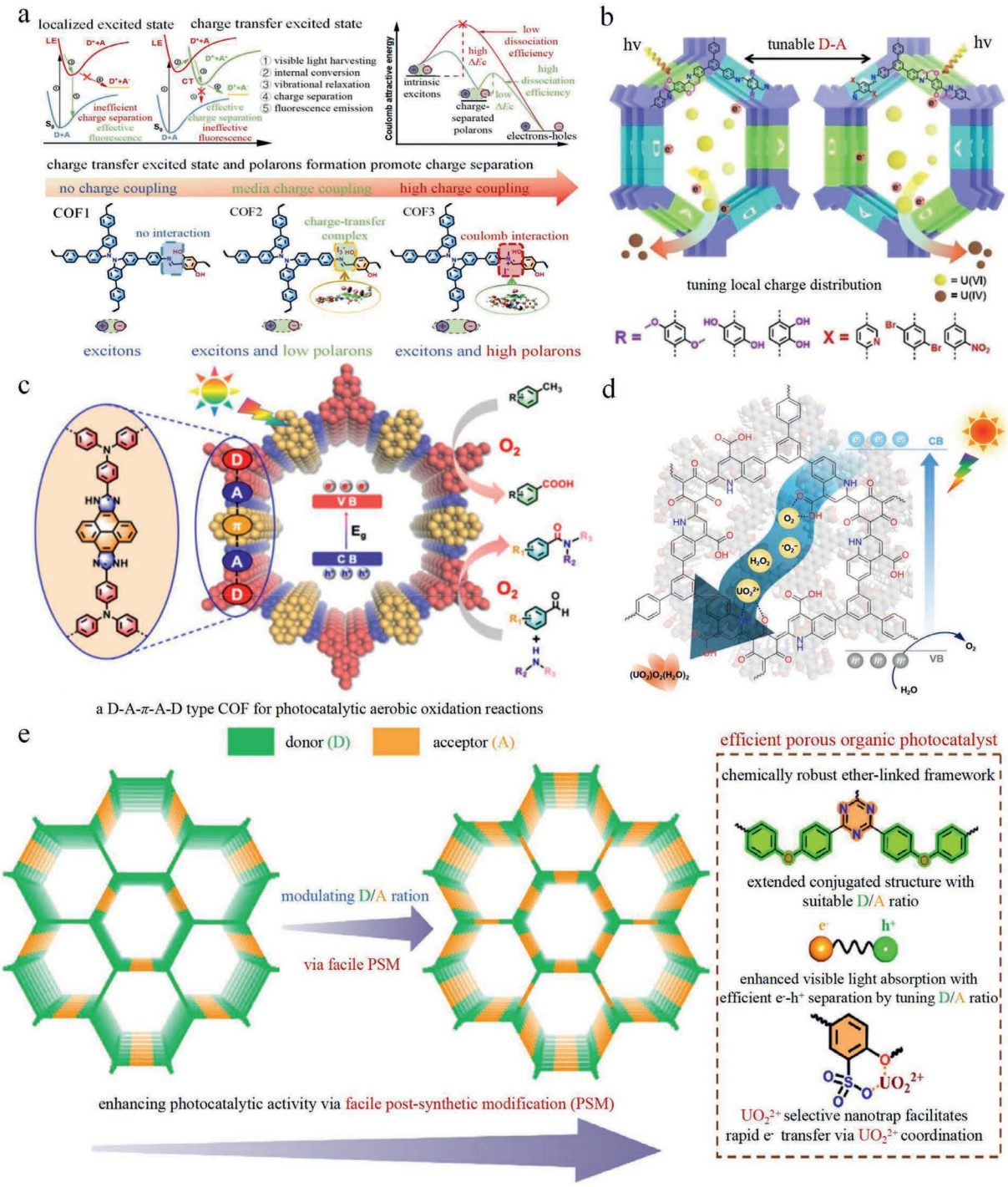


Fig. 2. (a) Schematic illustration of charge carrier separation, transfer and utilization by COF photocatalysts. Copied with permission [11]. Copyright 2026, Elsevier. (b) Schematic illustration for the construction of D-A systems through introducing electron donating and attracting groups on aromatic linkers in multivariate COF photocatalysts. Copied with permission [22]. Copyright 2023, Wiley. (c) Schematic diagram of the photocatalytic aerobic oxidation reaction of D-A-π-A-D type COFs. Copied with permission [29]. Copyright 2025, American Chemical Society. (d) Schematic illustration of photocatalytic uranium removal process *via* the multicomponent Doebner reaction. Copied with permission [30]. Copyright 2025, Elsevier. (e) Schematic presentation of the post-synthetic modification approach to enhance the photocatalytic activity of porous organic frameworks. Copied with permission [31]. Copyright 2025, Wiley.

absorption band was appeared obviously at 600–780 nm after doping anionic I^- and I_3^-/I_5^- onto the imine bonds of aniline-based COFs, which was significantly different with the undoped one (mainly at 200–580 nm) [11]. This strategy can simultaneously generate polarons and significantly inhibit the recombination of electron-hole (Fig. 2a). Lin *et al.* found that introducing ketone structure was helpful for light adsorption [19]. By converting the imine linkages into quinoline groups, the stability and photochemical properties of quinoline-linked COFs were obviously enhanced. The light absorption ranges from ~600 nm to ~700 nm, indicating that quinoline-linked COFs can increase the utilization range of light effectively [20]. Yang *et al.* investigated three imine-linked D-A COFs with different imine orientations (D-C=N-A or D-N=C-A). They demonstrated that D-N=C-A COFs showed significantly en-

hanced near-infrared light region absorption ability than D-C=N-A ones [21]. By incorporating a palladacycle into the COFs, the light absorption of azobenzene-based COFs was extended to near-infrared light region obviously.

For the electron-hole generation and separation, COFs also demonstrates excellent advantages and application prospects. Yang *et al.* found that the introduction of various electron-donating groups into D-A COFs could tune the local charge distribution and enhance charge carrier separation under visible light, and a high uranyl adsorption capacity of 8.02 mg/g/day from nature seawater was achieved without adding sacrificial reagents (Fig. 2b) [22]. The construction of ultrathin COF membranes can enhance exciton separation and improve photoactivity of COFs [23]. Interestingly, the designing of COFs heterojunctions is also an effective strategy to improve carrier separation [24].

The efficient transport and utilization of excited-state electrons is critical for the photoreduction and separation of uranyl. Our previous study showed that the excited electronic structure and charge transport pathways could be tuned by regulating the micro-local structure of COFs through oxygen containing functional groups [4]. Jiang and co-workers designed segregated columnar π-arrays COFs, which showed short D-A distance and could harvest singlet and triplet excitation energies. The π-columns in COFs can provide pathways for charge transport to the catalytic sites [25]. Qiu and co-workers observed that the construction of planar and continuous π-skeleton channels can reduce the binding energy of excitons (E_b) significantly and promote the separations of electron-hole pairs, thereby efficiently photo-reducing uranyl with extraction ability of 10.24 mg/g from the natural seawater [26].

Moreover, the photocatalytic activity of COFs can also be enhanced through multicomponent reactions or post-synthetic modification, providing robust support for photocatalytic uranyl reduction (Fig. 2c) [27–29]. Muzammil *et al.* reported that functionalized COFs synthesized *via* a multicomponent Doebner reaction were capable of generating $(UO_2)O_2(H_2O)_2$ under visible light through the reaction of *in situ* formed H_2O_2 with uranyl ions (Fig. 2d) [30]. Furthermore, a post-synthetic sulfonation strategy, achieved by tuning the donor-acceptor ratio, generated uranyl-specific nanotrapping sites while concurrently enhancing photocatalytic activity, thereby enabling the framework to realize highly efficient and ultrafast photoreduction of uranyl (Fig. 2e) [31]. Adsorption-photocatalysis strategy is efficient for electron transfer and direct utilization, as uranyl was the final electron "acceptor". The design of suitable uranyl photocatalytic sites is conductive to the efficient transmission and utilization of electrons.

To guide the rational design of high-performance COF–based photocatalysts for U(VI) extraction, it is useful to consider an integrated "adsorption-transport-reduction" framework (Fig. 3). In this context, adsorption serves primarily to enrich uranyl ions at catalytic interfaces and create a local environment where photocatalytic processes can act effectively. In adsorption-photocatalysis coupled systems, the improved uranyl adsorption ability can enhance uranyl aggregation and subsequent photocatalytic extraction efficiency [32]. Once uranyl species are concentrated at the surface, efficient transport of photoexcited charge carriers within the COFs becomes crucial. Structural features such as extended π–conjugation and D-A linkers have been shown to improve exciton migration and suppress charge recombination, leading to enhanced photoreduction performance of U(VI) by COFs [33]. Finally, the reduction step involves transfer of electrons to the adsorbed U(VI), yielding lower valence uranium species; this step depends on favorable band alignment and active photocatalytic sites that facilitate interfacial electron transfer and chemical conversion, as evidenced by studies in COFs and other porous frameworks where adsorbed U(VI) is efficiently reduced under light irradiation [34]. By viewing these stages as an interconnected sequence, the design of COFs photocatalysts can systematically balance enrichment, carrier transport, and photoreduction to achieve higher overall efficiency in uranium extraction.

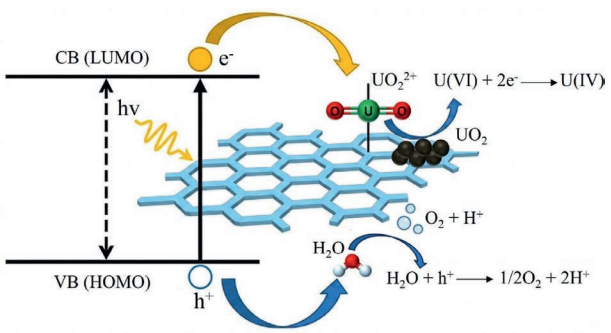


Fig. 3. Schematic illustration of U(VI) reduction by COFs during the photocatalytic process.

3. Perspectives

Although substantial progress has been made in elucidating uranyl reduction products and reaction pathways under well controlled or simplified photocatalytic systems, the detailed mechanisms operating under different photocatalytic conditions, especially in natural seawater and complex wastewater matrices remain incompletely understood. In this context, the integration of *in situ/operando* characterization techniques (such as XAFS, XRD, and Raman spectroscopy) with theoretical calculations is highly desirable to reveal the coupled "adsorption-photocatalysis" mechanisms and to track the evolution of uranium valence states and morphologies at the atomic and molecular levels.

The structural complexity and vast compositional diversity of COFs make traditional trial-and-error synthesis inefficient and often unpredictable. Artificial intelligence (AI)-driven approaches, particularly machine learning (ML) and deep learning (DL), are increasingly transforming this landscape by enabling data-driven predictions of structure-property relationships. Zhang *et al.* proposed an AI-assisted interactive evolutionary strategy that accelerates the discovery of novel covalent organic frameworks by integrating experimental exploration with iterative learning (Fig. 4). By embedding electronic structure descriptors and quantum mechanical understanding into the learning framework, this strategy moves beyond purely statistical intuition and establishes a chemistry informed materials design paradigm with enhanced predictive reliability and interpretability [35]. Wang *et al.* combined first-principles calculations with ML to predict the band-edge positions of COFs in photocatalytic processes, demonstrating high accuracy in experimental validation [36]. Building on this foundation, future studies may integrate density functional theory (DFT) datasets with ML models to predict structural features, including π-conjugation length, D-A motifs, and heteroatom composition, and subsequently infer key electronic properties of COFs, such as bandgap energies and band-edge positions, thereby enabling rapid screening of COFs with suitable band alignments for photocatalytic uranyl reduction. Meanwhile, ML-assisted structure–property analysis can correlate local coordination environments and functional groups (such as carbonyl- or nitrogen-containing moieties) with computed uranyl binding energies, thereby identifying and optimizing favorable uranyl coordination sites. These AI-driven approaches provide a powerful and scalable framework for accelerating the rational design of high-performance COFs for photocatalytic uranium extraction. However, the limited size, quality, and consistency of datasets can constrain model generalization, and pre-

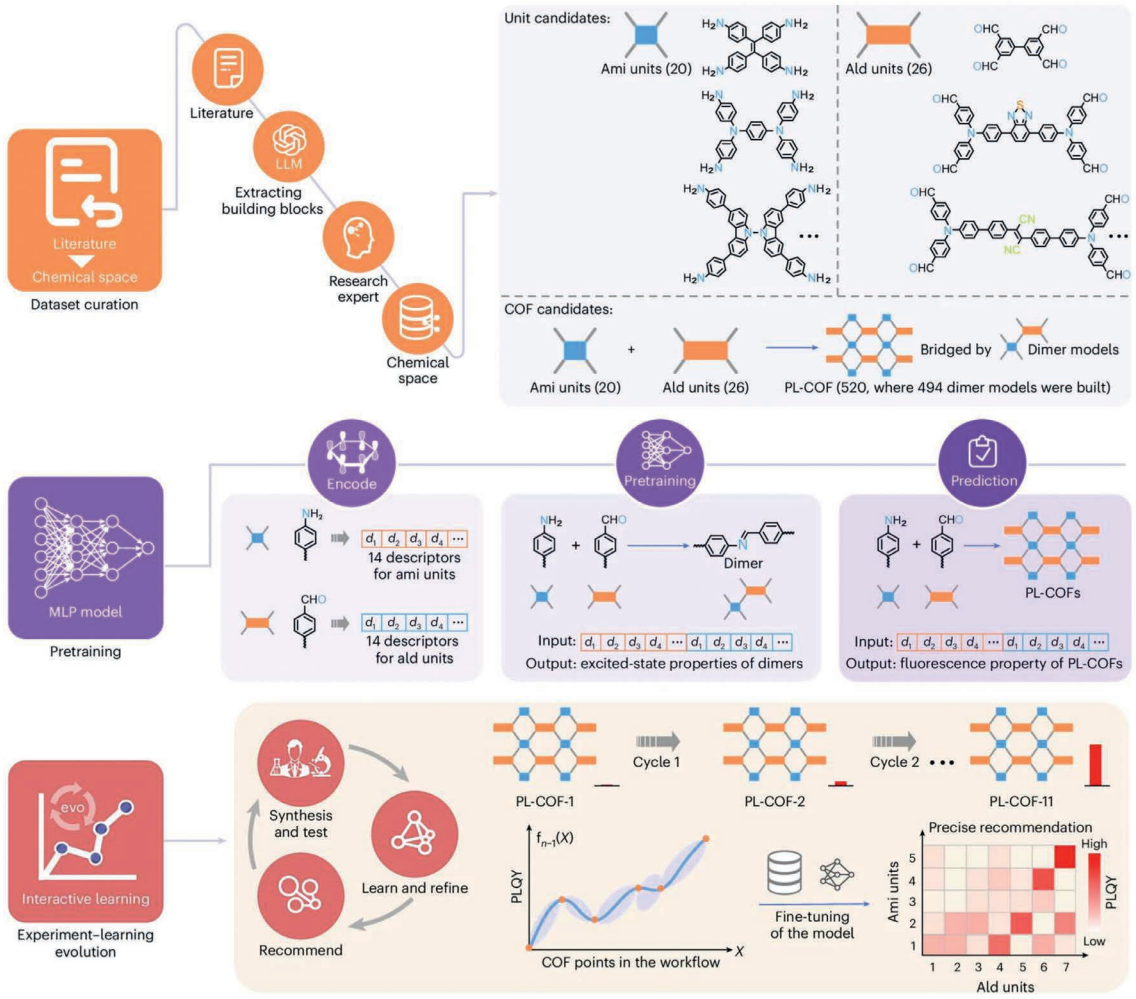


Fig. 4. Schematic illustration of the AI-assisted workflow for discovering novel COFs. Copied with permission [35]. Copyright 2025, Springer Nature.

dictions often require experimental validation. Future efforts integrating standardized datasets, first-principles calculations, and machine learning are expected to accelerate the rational design of high-performance COFs for uranium extraction.

For practical applications of COFs in photocatalytic uranyl adsorption and extraction, several issues still need to be considered, including material cost, long term stability in complex aqueous environments, continuous operation, and recyclability. These challenges are largely related to the fact that most COFs are obtained as fine powders, which are difficult to handle in large scale systems and are not well suited for continuous extraction processes. In addition, powder catalysts often suffer from limited mass transfer, catalyst loss during operation, and difficulties in recovery and reuse. Constructing COFs into macroscopic forms such as aerogels, foams, films, or fibers provides a practical way to address these problems. COF aerogels and foams contain interconnected porous channels that can promote mass transfer of uranyl species and improve light penetration during photocatalysis. In contrast, COF films or fibrous membranes can be directly integrated into continuous flow or fixed bed reactors, which is beneficial for long term operation and repeated use without complicated separation steps. Moreover, forming composites can improve the mechanical strength and structural stability of COFs, which is important for their application in seawater or wastewater systems. With further progress in material processing and synthesis, together with the assistance of AI-guided molecular design, it is expected that COFs with suitable structures and good visible-light response can be developed for more efficient photocatalytic uranium extraction. Although challenges remain, large scale and low cost uranium extraction from seawater may become feasible in the future.

CRediT authorship contribution statement

Zhongshan Chen: Writing – original draft, Investigation, Funding acquisition, Data curation, Conceptualization. **Jianen Li:** Writing – original draft, Investigation, Data curation, Conceptualization. **Qiao Ma:** Writing – original draft, Investigation, Data curation. **Xishi Tai:** Writing – review & editing, Conceptualization. **Jiehong Lei:** Writing – review & editing, Conceptualization. **Xiangke Wang:** Writing – review & editing, Supervision, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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