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Recent advances in the construction of functionalized covalent organic frameworks and their applications to sensing

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ABSTRACT

Covalent organic frameworks (COFs), as an emerging class of porous crystalline polymers, are built by the combination of the light elements through the strong covalent bonds. In the past decade, COFs have been reported to show plenty of unique properties (such as ordered channels, large specific surface area, highly tunable porosity, optional building blocks, predictable and stable structure, and abundant functional groups), and have been widely applied in multiple fields. Recently, to further improve the potential performances of COFs and extend their applicability, a number of COFs with various functionalities have been successfully developed through the functionalization modification. In this review, we summarized the advanced design and construction of functionalized COFs, including COFs with post-synthetic modification, COFs-based composites (e.g. COFs-metal nanoparticles composites, COFs-metal oxide nanoparticles composites, COFs-MOFs composites, and COFs-enzyme composites), and molecularly imprinted COFs. Impressively, the applications of functionalized COFs to sensing also have been comprehensively summarized, including colorimetric sensing, fluorescent sensing, electrochemical sensing, and other sensing (such as quartz crystal microbalance (QCM) sensing, photoelectrochemical sensing, and humidity sensing). In the end, future opportunities and challenges in this promising field are tentatively proposed.

1. Introduction

Covalent organic frameworks (COFs), as a kind of emerging porous crystalline polymers, which are built by the combination of the light elements (e.g. B, C, Si, N, and O) through the strong covalent bonds (e.g. B–O, C–N, C=N, and C=C–N) with atomically precise spatial assembly, discrete pores, and rigidly ordered organic building blocks (Côté et al., 2005; Ding and Wang, 2013; Feng et al., 2012; Yuan et al., 2019). The geometries of the COFs are determined by the selection, symmetry, size, and connectivity of linkers and building blocks (Chen et al., 2019b). Since the two dimensional (2D) COFs (COF-1 and COF-5) were successfully synthesized by Yaghi's group for the first time in 2005, COFs have attracted much attention in recent years due to their great development potential, and a great many novel COFs have been developed(Côté et al., 2005; Lin et al., 2016; Lv et al., 2019; Yuan et al., 2019). Based on the dimension of the building blocks, the COFs can be classified into 2D COFs and 3D COFs (Díaz and Corma, 2016; El-Kaderi et al., 2007). The 3D COFs with microporosity can be prepared using associated rigid spyrocyclic, aromatic or arylene linkers through self-condensation processes. The 3D COFs show poor porous homogeneity and low crystallinities, and same as hyperbranched or conjugated microporous polymers. Compared to 3D COFs, 2D COFs are easier to be synthesized due to their simple structure. The 2D COFs are formed through ordered piled layers, and can be prepared based on the ability of rigid organic units to generate the individual nanosheets (Díaz and Corma, 2016). In the past decade, the COFs have been reported to show some unique properties, such as ordered channels, large specific surface area, highly tunable porosity, optional building blocks, predictable and stable structure, and abundant functional groups, which endow them with outstanding performances for multiple fields, such as energy

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Abbreviations

AMP	ampicillin
Bd	benzidine
Bth	benzene-1,3,5-tricarbohydra-zide
COFs	covalent organic frameworks
COF-HQ	HQ-functionalized COF
CEES	2-chloroethyl ethyl sulfide
CNs	carbon nanodots
cTnI	cardiac troponin I
DMTP	2,5-dimethoxyterephthaldehyde
Dma	2,5-dimethoxyterephthal-aldehyde
Dha	5-dihydroxyterephthalaldehyde
dsDNA	double-stranded DNA
EB	ethidium bromide
ET	electron transfer
ENR	enrofloxacin
EGFR	epidermal growth factor receptor
Fe–COF	Fe-porphyrin-based COF
Fe^{3+}	Fe (III) ion
GS-COF	graphene-synergized 2D COF
GOx	glucose oxidase
GCE	glassy carbon electrode
HBI	2-(2,4-dihydroxyphenyl)-benzimidazole
HRP	horseradish peroxidase
IFE	inner filter effect
LOD	limit of detection
LSPR	localized surface plasmon resonance
MNPs	metal nanoparticles
MICOFs	molecularly imprinted COFs
MIOP	molecularly-imprinted optopolymer
MCF-7	michigan cancer foundation-7
NSAIDs	nonsteroidal anti-inflammatory drugs
PVP	polyvinylpyrrolidone
PDA	polydopamine
PI-CONs	PI covalent organic nanosheets
Pd	palladium
Pa-1	paraphenylenediamine
PI-COF	polyimide COF
p–COF–10	pyridine-modified COF-10
p-COF	porphyrin-based COF
QCM	quartz crystal microbalance
QCA	quinoxaline-2-carboxylicacid
SMR	sulfamerazine
SERS	surface-enhanced Raman scattering
SPR	surface plasmon resonance
Tb	1,3,5-triformylbenzene
Tp	1,3,5-triformylphloroglucinol
TYM	tyramine
TNP	2,4,6-trinitrophenol
TMB	3,3′,5,5′-tetramethylbenzidine
TAPB	1,3,5-tris(4-amino-phenyl)benzene
VOCs	volatile organic compounds
4-EG	4-ethylguaiacol

conversion and storage(Xiang et al., 2016), separation(Yuan et al., 2019), gas adsorption(Das et al., 2017; Huang et al., 2015), analysis (Chen et al., 2019b), catalysis(Li et al., 2016b; Xu et al., 2015; Zhi et al., 2018), and sensing(Wu et al., 2019). In addition, tunable porosity, π conjugation, large surface area, and unique photoelectric properties of COFs have enabled them to serve as an extremely promising platform for biomedical field, including drug delivery, sterilization, bioimaging, theranostic, etc.(Hynek et al., 2018; Lohse and Bein, 2018; Song et al., 2019). Recently, to improve the potential performances of COFs and extend their applicability, plenty of COFs with various functionalities are successfully developed through the functionalization modification (Babu et al., 2019; Chen et al., 2018; Das et al., 2017; Gontarczyk et al., 2017; Hynek et al., 2018; Ji et al., 2018a). The post-synthetic modification of COFs is proposed for introduction of various functional groups or side groups, which allows COFs with improved functions (Huang et al., 2016; Lohse and Bein, 2018). For example, COFs functionalized by amine are used to removal the perfluorinated alkyl substances and ammonium perfluoro-2-propoxypropionate (GenX) from water (Ji et al., 2018b). Diacetylene functionalized β-ketoenamine COFs can be used for the photocatalytic hydrogen production (Pachfule et al., 2018). It is reported that some molecularly imprinted polymers-functionalized COFs (MICOFs) also have been successfully developed for separation (Ji et al., 2018a), sensing(Zhang et al., 2019a), molecular recognition(Yan et al., 2018), and so on(Zhang et al., 2019b). In addition, to endow COFs with various functionalities, some COFs-based composites have also been prepared through the integration of COFs with metal nanoparticles (MNPs), metal oxide nanoparticles, metal-organic frameworks (MOFs), biological enzymes, and so on. For example, the Pd(II)-containing COFs (Pd/COF-LZU1) with excellent catalytic activity are successfully synthesized through a simple post-treatment method for the catalysis of the Suzuki-Miyaura coupling reaction(Ding et al., 2011). Through the rapid room-temperature synthesis, the core-shell structured magnetic COF composites (Fe_3O_4@TbBd) are prepared using benzidine (Bd) and 1, 3, 5-triformylbenzene (Tb) as building blocks and monodisperse Fe_3O_4 NPs as the magnetic core, respectively. The prepared Fe_3O_4@TbBd can be applied in the selective enrichment of hydrophobic peptides with a simultaneous exclusion of proteins (Gao et al., 2017). To obtain a novel porous hybrid material as a highly effective visible-light-driven photocatalyst, the MOFs integrated with COFs can produce a novel type of MOF@COF core-shell hybrid material (NH_2-MIL-68@TPA-COF hybrid material) for the degradation of rhodamine B (Peng et al., 2018). To obtain the COFs and enzyme biocomposites with the enhanced activity, the monomers bearing specific functional groups are used to tune the pore environment of COFs for the improvement of their compatibility with enzymes, and the enzyme active site orientation is modulated by the designed interactions between COFs and enzymes (Sun et al., 2018b). Based on their various functionalities, the functionalized COFs have been extensively applied in multiple fields, such as catalysis, biomedicine, drug delivery, energy conversion and storage, gas storage and separation, and sensing(Chakraborty et al., 2019; Chen et al., 2019b; Guo et al., 2019; Huang et al., 2016; Liu et al., 2019b; Qiu et al., 2019; Wang et al., 2018a).

In previous reports, the COFs and their applications to analytical chemistry(Chen et al., 2019b), heterogeneous catalysis(Zhi et al., 2018), separation science(Qian et al., 2018), sample pretreatment(Li et al., 2018b), and biomedical(Lohse and Bein, 2018) have been reviewed. However, the design and construction of functionalized COFs and their sensing applications are rarely summarized. In this review, we comprehensively summarized the advanced design and construction of functionalized COFs, including COFs with post-synthetic modification, COFs-based composites (e.g. COFs-metal nanoparticles composites, COFs-metal oxide nanoparticles composites, COFs-MOFs composites, and COFs-enzyme composites), and molecularly imprinted COFs (MICOFs). Impressively, the applications of functionalized COFs to sensing also have been comprehensively summarized, including colorimetric sensing, fluorescent sensing, electrochemical sensing, and other sensing (such as QCM sensing, photoelectrochemical sensing, and humidity sensing).

2. Construction strategies of functionalized COFs

2.1. COFs with post-synthetic modification

Based on their regular pores with a defined pore environment, COFs can be decorated with various linkers during the synthesis process to change the pore environment. Nevertheless, the variation ranges of COFs pore environment often suffer a great limit. The post-synthetic modifications of the COFs as an alternative strategy is established for introduction of functional groups or large side groups, which can endow COFs with some distinctive functions and greatly broaden their applications (Beuerle and Gole, 2018; Lohse and Bein, 2018; Segura et al., 2019). In a previous review article, Segura et al. have systematically reviewed post-synthetic modification of COFs, which mainly focused on the post-synthetic modification mechanism and approaches (Segura et al., 2019). While in the present review, we mainly focused on summarizing the advanced functionalized COFs, including COFs with post-synthetic modification, COFs-based composites, and molecularly imprinted COFs. Moreover, the applications of advanced functionalized COFs in the field of sensing have been systematically summarized.

2.1.1. COFs with post-synthetic intercalation and complexation

Based on the previous reports, it is found that the post-synthetic intercalation and complexation way have been widely applied in

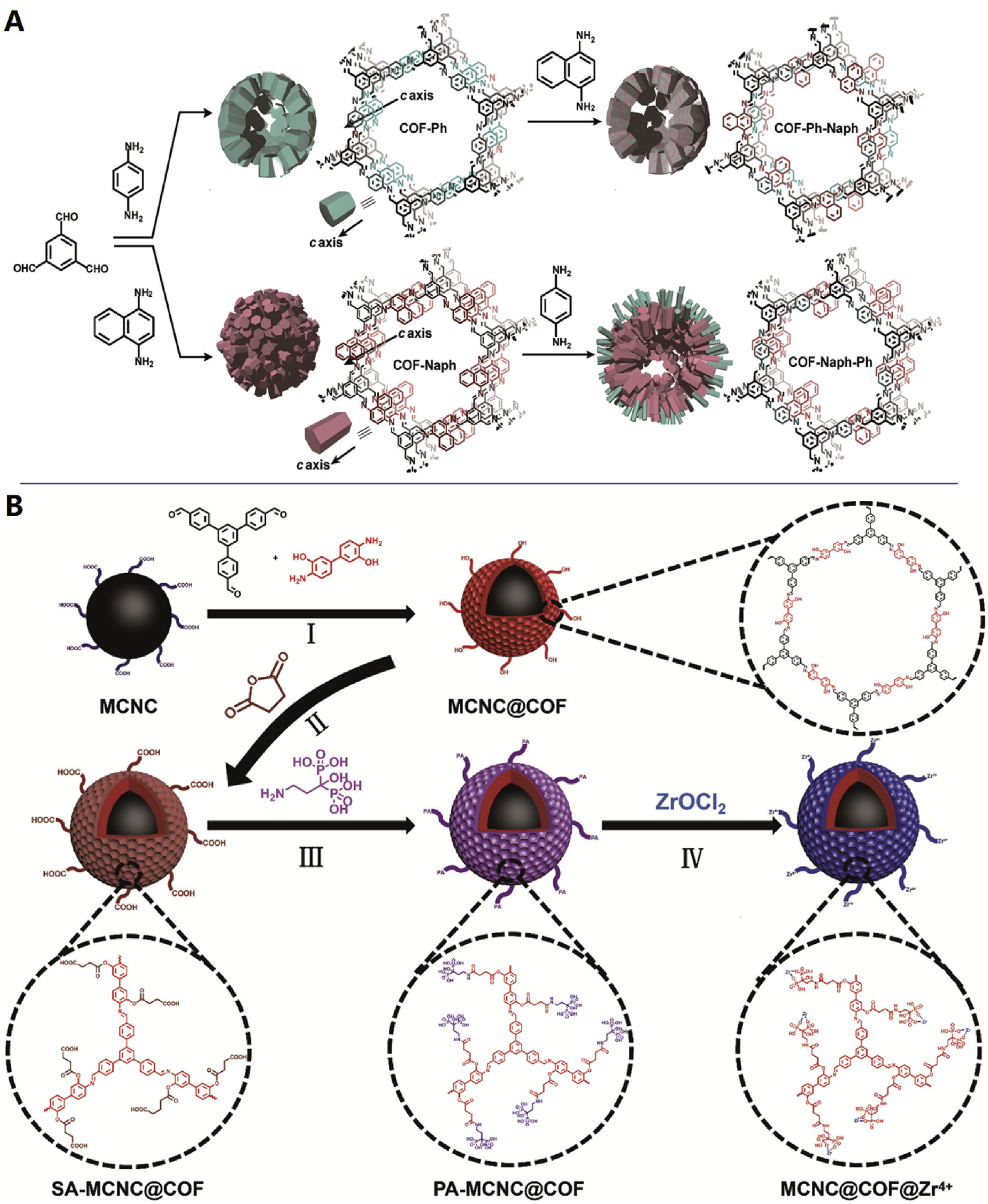

Fig. 1. (A) Synthesis of COF-Ph, COF-Naph, COF-Ph-Naph, and COF-Naph-Ph by a postsynthetic process and representation(Zhang et al., 2018a). (B) Representation of the strategy for preparing Zr^{4+}-immobilized magnetic COFs through sequential postsynthetic modifications(Gao et al., 2019).

improvement of COF performances (Hu et al., 2019; Lohse and Bein, 2018). For example, Du et al. prepared pyridine-modified COF-10 (p–COF–10) through the postsynthetic introduction of pyridine into the pores of the as-synthesized COF-10. Compared to the un-modified COF-10, the as-prepared p–COF–10 exhibited an enhanced stability in humid air (Du et al., 2012). To obtain a novel solid-phase extractant with an extremely efficient sorption ability for the uranium (VI), Li et al. modified the as-prepared COF–COOH with the previously fabricated 2-(2,4-dihydroxyphenyl)-benzimidazole (HBI) for preparation of COF-HBI(Li et al., 2015). In another study, by utilizing the dynamic covalent chemistry-based postsynthetic method, Zhang et al. prepared the 2D imine-bonded COFs with distinct aromatic groups, and finally obtained a heterogeneously core-shell hollow structure and a homogeneously mixed-linker structure through the conformity of the postsynthetic reaction reactivity (Fig. 1A)(Zhang et al., 2018a). In another study, Chen et al. successfully selected and introduced a pH-sensitive fluorescent chemical group (8-hydroxyquinoline (HQ)) into the channels of the 2D COF to prepare the HQ-functionalized COF (COF-HQ) for the dual-mode pH sensing(Chen et al., 2018). To achieve high capacity and rapid uptake for dye pollutants, Biradha's group rationally designed and successfully obtained three luminescent triazene-based COFs (COFs 1–3) functionalized with azine and imine by forming imine bonds through the condensation between linear diamines and flexible tripodal tri-aldehydes (TFPT)(Konavarapu and Biradha, 2019). Based on the vapor phase infiltration of volatile organometallic precursors, Fischer's group studied the organometallic host-guest chemistry of COFs. The intercalation of ferrocene molecules into COF-102 in an oriented way was driven through the host-guest interactions and replicated the symmetry of framework. The COFs showed good stability upon loading with the ferrocene molecules, and also retained their structure upon the incorporation of $[Ru(cod)(cot)]_2$ or cobaltocene(Kalidindi et al., 2011). In addition, by the post-synthetic introduction of metal ions into COFs,

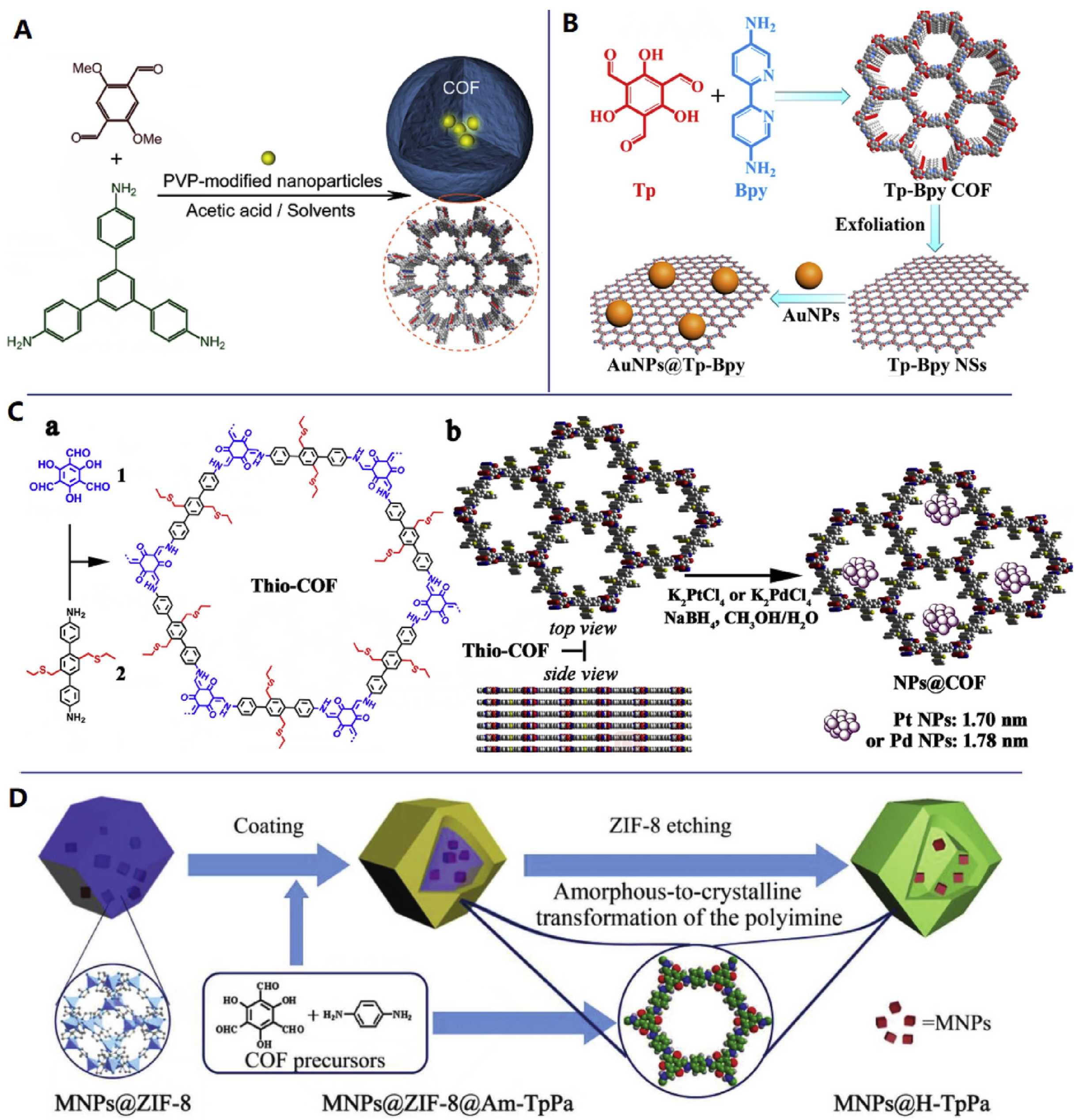

Fig. 2. (A) Schematic illustration of the encapsulation of PVP-modified nanoparticles in the 2D TAPB-DMTP-COF(Shi et al., 2017). (B) Synthesis method of AuNPs@Tp-Bpy nanocomposite(Cui et al., 2019b). (C) Synthesis of Thio-COF and schematic representation of the synthesis of Thio-COF supported PtNPs@COF and PdNPs@COF(Cui et al., 2019b). (D) Schematic illustration of the encapsulation of MNPs in a COF nanocage(Cui et al., 2019a).

the metal centers could fixed at the specific positions in the porous COF scaffold, resulting in separate and defined reaction centers for excellent sorption and catalysis applications(Lohse and Bein, 2018). For example, Ding and co-workers for the first time reported a metal complex in the COF (Pd/COF-LZU1) with excellent activity for the catalysis of the Suzuki-Miyaura coupling reaction. Due to the two-dimensional eclipsed layered-sheet structure of imine-linked COF-LZU1, it could feasibly incorporate with metal ions. On this basis, the Pd/COF-LZU1 was prepared by the post-synthetic modification of obtained imine COF LZU-1 with $Pd(OAc)_2$ through a simple post-treatment (Ding et al., 2011). By using vanadyl acetylacetonate, Vardhan et al. post-synthetically modified the 2D COFs and obtained vanadium-decorated COFs (VO-TAPT-2, 3-DHTA COFs) with large pores, eclipsed stacking structure, hydroxyl functionalities, and highly thermal and chemical stability for the sulfide oxidation and Prins reaction(Vardhan et al., 2019). In a recent study, through sequential post-synthetic functionalization, Gao et al. innovatively synthesized a novel controllable core-shell structured Zr^{4+}-immobilized magnetic COFs (MCNC@COF@Zr^{4+}) by employing the MCNC with high-magnetic-response as core and the Zr^{4+} ion-functionalized COFs as the shell, respectively (Fig. 1B). Moreover, the obtained MCNC@COF@Zr^{4+} showed higher Zr^{4+} ion loading amount, good thermal and chemical stability, regular porosity with large surface areas, and strong magnetic responsiveness (Gao et al., 2019).

2.1.2. COFs with covalent post-synthetic modification

Through covalent post-synthetic modification, Nagai et al. for the first time achieved the pore surface engineering in COFs via the formation of covalent bond. In this study, the different fractions of azide functionalized BDBA were introduced into the boronic ester frameworks produced by the BDBA and Ni-phthalocyanine catechol or BDBA and HHTP, respectively. The azide moieties could make a reaction with different alkynes through Cu^{+}-catalyzed click reaction after lattice formation. In addition, the amount of groups by post-synthetic introduction could be controlled through the tune of incorporated azide percentage (Nagai et al., 2011). Utilizing the same way, Chen et al. combined the functionalized fullerene with the pore walls of boronic ester phthalocyanine COFs through the covalent binding (Chen et al., 2014). By using the COF (COF-102), Bunck et al. developed a new tandem truncation-functionalization approach to establish the covalent docking sites in COFs. The full functionalization of the incorporated allyl groups could be achieved via the thiol-ene coupling reaction between allyl groups and propanethiol (Bunck and Dichtel, 2013). In a previous literature, Lohse et al. reported the post-synthetic modification of the stable COF (β-ketoenamine TpBD $(NH_2)_2$), with the integration of primary amine groups into their pore walls. The as-prepared functionalized COF showed the high stability under acidic conditions, which could be used for the liquid-phase adsorption of lactic acid (Lohse et al., 2016). Recently, Zhang et al. successfully developed two chiral functional COFs (TPB2-COF and Tfp2-COF) using chiral organic building blocks through a solvo-thermal strategy. The prepared Tfp2-COF and TPB2-COF could be used as green, sustainable, efficient, lower cost, and reusable heterogeneous asymmetric organocatalysts for the catalysis of multiple asymmetric organic reactions (Zhang et al., 2019c).

2.2. COFs-based composites

COFs have been considered to be the excellent host matrices for plenty of functional species due to their open porous structures and good stability. Integration of functional materials (e.g. MNPs, metal oxide NPs, MOFs, biological enzymes, and quantum dots) with COFs is a promising strategy to enhance the performances of COFs and extend their applications. To date, a large number of COFs-based composites have been successfully developed, including COFs-MNP composites, COFs-metal oxide NP composites, COFs-MOFs composites, COFs-enzyme composites and so on. By the integration between the COFs and functional materials, COFs can be endowed with some enhanced or new functions, such as higher catalytic activity, good magnetism, stronger fluorescence, and enhanced adsorption capacity(Li et al., 2019a; Wang et al., 2018a; Xia et al., 2018; Zhang et al. 2018b, 2018e, 2019b).

2.2.1. COFs-metal nanoparticles composites

Metal nanoparticles (MNPs) have attracted extensive interests in the past few years due to their outstanding catalytic properties. Unfortunately, the aggregation of MNPs (e.g. AuNPs, PtNPs, and PdNPs) is easily formed owing to their high surface energy, which leads to their reduced catalytic activity and difficult long-term storage. Therefore, the accessibility and dispersibility of MNPs are of importance for improving their catalytic activity (Cui et al., 2019a; Lu et al., 2017; Su et al., 2019). Recently, utilizing the open porous structures and excellent stability of COFs, some MNPs such as AuNPs, PtNPs, and PdNPs have been integrated with COFs to produce new COFs-MNPs composites. By the combination of COFs and MNPs, COFs-MNPs composites obtained the outstanding catalytic activity of MNPs and the merits of COFs (Liu et al., 2016; Zhang et al., 2018e). For example, Shi et al. successfully prepared AuNPs/TAPB-DMTP-COF composites (Fig. 2A) by incorporating AuNPs with catalytic activity and different shapes, sizes, and contents in 2D COFs produced from the condensation of 2,5-dimethoxyterephthaldehyde(DMTP) with 1,3,5-tris (4-amino-phenyl)benzene (TAPB). The encapsulation of AuNPs into 2D COFs was achieved based on the polyvinylpyrrolidone (PVP) functionalized AuNPs and then their adsorption onto the surface of COFs polymeric precursors. Moreover, the pore structures, crystallinity, and thermal stability of 2D COFs were obviously changed by the incorporation of AuNPs. The as-prepared AuNPs/TAPB-DMTP-COF composites showed highly open mesopores, large surface areas, and excellent recyclable catalytic activity for the reduction of 4-nitrophenol (Shi et al., 2017). Using the strong electrostatic interaction between AuNPs and the unsaturated amine groups of TAPB-DMTP-COFs, Zhang et al. employed COFs as the host matrix to immobilize AuNPs onto the surface of TAPB-DMTP-COFs under vigorously stirring for the preparation of AuNPs-doped COF composites (Zhang et al., 2018c). Recently, Gu et al. employed the porous light-weight material COFs (CTpBD) and AuNPs to prepare the COFs-AuNPs composite for the construction of QCM sensor (Gu et al., 2019). In a very recent study, Cui et al. successfully prepared bipyridine-containing COF nanosheets (Tp-Bpy NSs), which possessed plenty of nitrogen-containing functional groups and the regular pore structure. On the basis of abundant active sites of obtained COF nanosheets, AuNPs@Tp-Bpy could be synthesized by in situ generation of AuNPs on the COF nanosheets (Fig. 2B). Moreover, the catalytic activity, stability, and dispersibility of AuNPs could be significantly enhanced by the anchor of AuNPs onto COF nanosheets via coordination bonds (Cui et al., 2019b). In addition to COFs-AuNPs composites, COFs-PtNPs composites and COFs-PdNPs composites also have been successfully developed (Yao et al., 2018). For example, Lu et al. rationally designed and successfully prepared the thioether-containing COFs as promising templates for the immobilization of ultrafine PtNPs and PdNPs inside the cavity of COFs (Fig. 2C). The PtNPs and PdNPs immobilized onto the COFs exhibited a high capacity and narrow size distribution. Under low catalyst loading and mild conditions, the obtained COFs-supported ultrafine PdNPs and PtNPs possessed outstanding catalytic performance in Suzuki-Miyaura coupling reaction and nitrophenol reduction, respectively (Lu et al., 2017). In a recent study, employing MNPs@ZIF-8 core-shell nanostructures as the self-template, Cui and co-workers designed the yolk-shell MNPs@COF nanoreactors with high catalytic activity (Fig. 2D). In a mildly acidic solution, the COF shell was produced by the amorphous-to-crystalline transformation process of the polyimine shell. The void space between the COF shell and the MNP core was generated with the in situ etching of ZIF-8. Owing to the protection of H-TpPa shell, the plenty of ligand-free PdNPs were confined inside of hollow H-TpPa. The as-prepared Pd@H-TpPa yolk-shell nanocages with the permeable TpPa shells and active PdNP cores showed excellent catalytic activity and good stability for the reduction of 4-nitrophenol via $NaBH_4$ under the room temperature condition (Cui et al., 2019a).

2.2.2. COFs-metal oxide nanoparticle composites

Recently, the integration of COFs with Fe_3O_4 for the preparation of magnetic COF composites with the advantages of outstanding extraction and fast magnetic separation has attracted much attention (Deng et al., 2019a, b; Li et al., 2018a; Li et al., 2018d; Shi et al., 2018; Zhang et al., 2019d). For example, to effectively enrich and detect the trace polycyclic aromatic hydrocarbons, Li et al. successfully synthesized novel core-shell nanostructure magnetic COF hybrid microspheres (Fe_3O_4@COF-(TpBD)) with the larger specific surface area, uniform morphology, retention of colloidal nanosize, supermagnetism, and higher porosity(Fig. 3A)(Li et al., 2018c). Employing the TbBd as an outer COF shell, polydopamine (PDA) as the hydrophilic middle layer, and Fe_3O_4 NPs as the magnetic core, Yan et al. facilely synthesized a novel functionalized COF-derived material (Fe_3O_4@PDA@TbBd) for efficient determination of phthalic acid esters (PAEs) (Fig. 3B)(Yan et al., 2018). Zhai et al. for the first time developed an immunoaffinity probe (Fe_3O_4@TpBD-DSS-Ab-MEG) using magnetic COFs with the anchored antibodies (Fig. 3C). The as-prepared Fe_3O_4@TpBD-DSS-Ab-MEG possessed the merits from the Fe_3O_4 cores and COF shells, showing strong magnetic responses, highly thermal and solvent stability, and ordered active groups (Zhai et al., 2019). Jiang et al. innovatively prepared a new solid phase extraction adsorbent using aptamer-functionalized magnetic COFs, which showed the superparamagnetism, porous structure, high surface area, and highly specific affinity to target (Jiang et al., 2018). In addition to magnetic COFs-Fe_3O_4 composites, other COFs-metal oxide nanoparticle composites have also been successfully prepared. For example, Chakraborty et al. developed COF-based Cu catalyst by growing Cu/Cu_2O NPs (2–3 nm) onto the COFs produced by the link between the phenolic trialdehyde and triamine via Schiff bonds. The NPs were restricted to be small sizes (about 2–3 nm) by the micropores on COFs, and the substrates were anchored by the pore walls modified with positioned hydrogen-bonding phenolic groups through hydrogen-bonding. The $[Cu_{cluster}Cl_2]^{2-}$ type reactive intermediates were stabilized by the basic pyridyl sites that served as cationic species. The prepared composite catalyst exhibited high catalytic activity for Glaser-Hay heterocoupling reactions, and showed the elevated selectivity for heterocoupling products (Chakraborty et al., 2019).

2.2.3. COFs-MOFs composites

MOFs as a type of crystalline hybrid materials constructed from metal ions or metal cluster nodes interconnected through organic ligands, which have attracted a great many research interests due to their unique properties and extremely promising applicability (Feng et al., 2018; Lohse and Bein, 2018; Lu et al., 2018; Zhao, 2016). Based on their outstanding properties of large surface areas, tunable structures, and highly chemical and thermal stabilities, which have showed potential applications in multiple fields, such as catalysis, gas storage and separation, nonlinear optics, drug delivery, energy conversion, and sensing (Lu et al., 2018; Zhou et al., 2019). To improve the potential performances of COFs and broaden their applicability, the integration of MOFs with COFs as a promising strategy can endow COFs with much more outstanding performances (He et al., 2019a; Li et al., 2019a; Zhou

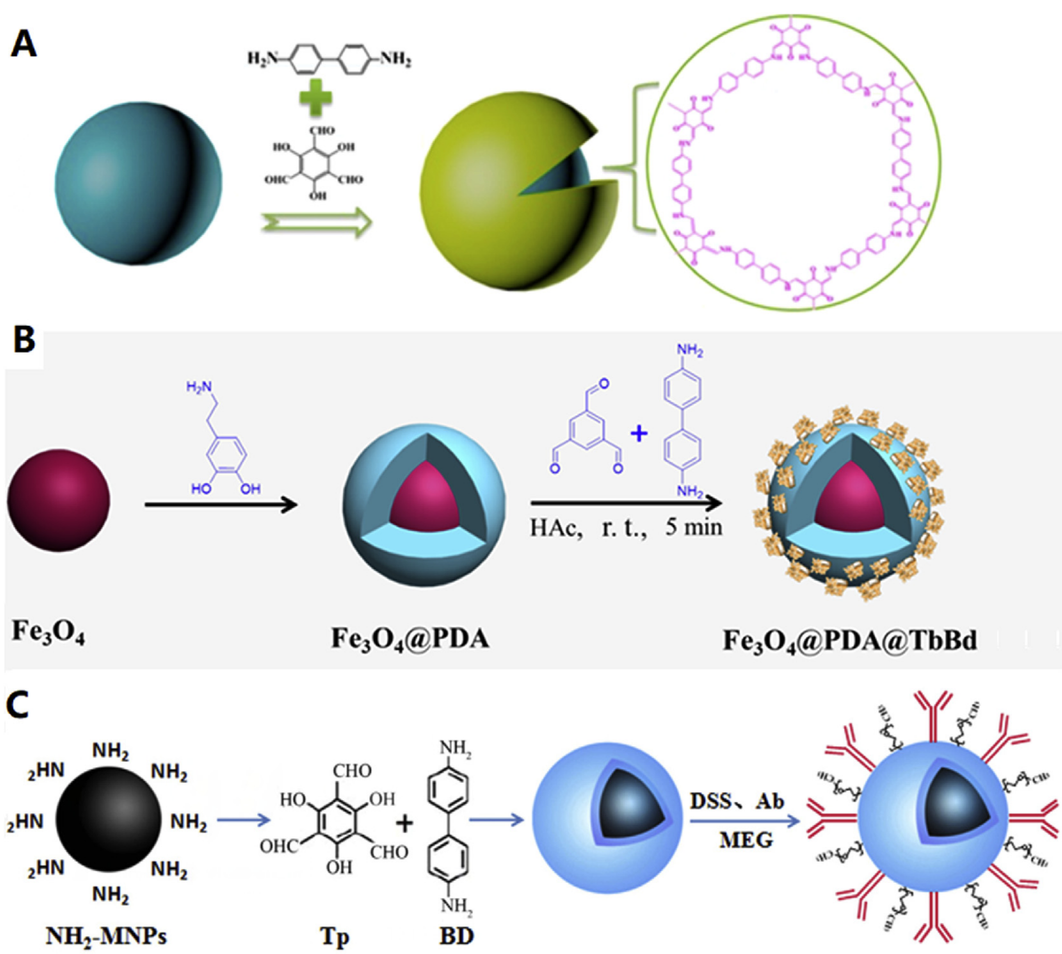

Fig. 3. (A) Schematic fabrication process of Fe_3O_4@COF-(TpBD) (Li et al., 2018c). (B) Synthetic strategy of Fe_3O_4@PDA@TbBd(Yan et al., 2018). (C) Synthetic route of Fe_3O_4@TpBD-DSS-Ab-MEG immunoprobe(Zhai et al., 2019).

et al., 2019). Recently, COFs-MOFs composites have been developed and widely applied in gas separation, photocatalysis, separation, biosensing, electrocatalyst and so on(Dutta et al., 2015; Li et al., 2019a; Liu et al., 2019c; Sun et al., 2018a). For example, Matzger et al. innovatively integrated the construction principles of COFs and MOFs into a framework. In this study, the triangular carboxylic acids were applied to form the MOFs with amino functional group. The frameworks with the coordinative linkages and covalent imine bonds were formed through the reaction between the dialdehydes and Zn $(NO_3)_2{\cdot}6H_2O$(Dutta et al., 2015). In another study, Nguyen et al. developed a new crystalline material with a 2D structure by the combination of COF and MOF nanostructure, which involved in situ generation of amine-functionalized hexameric Ti(IV) clusters $(Ti_6O_6(OCH_3)_6(AB)_6)$ connected with the benzene-1,4-dialdehyde through imine condensation reactions(Nguyen et al., 2016). By integrating COFs with MOFs, Peng et al. developed a novel type of MOF@COF core-shell hybrid material (NH_2-MIL-68@TPA-COF) with hierarchical pore structure and high crystallinity. The as-synthesized NH_2-MIL-68@TPA-COF hybrid material served as an excellent visible-light-driven photocatalyst for degrading the rhodamine B (Peng et al., 2018). To develop a sensitive electrochemical aptasensor for the detection of oxytetracycline, Zhou et al. for the first time rationally designed and successfully prepared new kinds of nanohybrids of COFs and Ce-MOFs as the non-label bioplatforms (Fig. 4A). The Ce-MOF@COF nanohybrids were synthesized via the addition of different dosages of COFs prepared by the polycondensation between cyanutic acid-monomers and melamine into the fabrication system of Ce-MOFs. Moreover, the Ce-MOF@ COF nanohybrids possessed their original

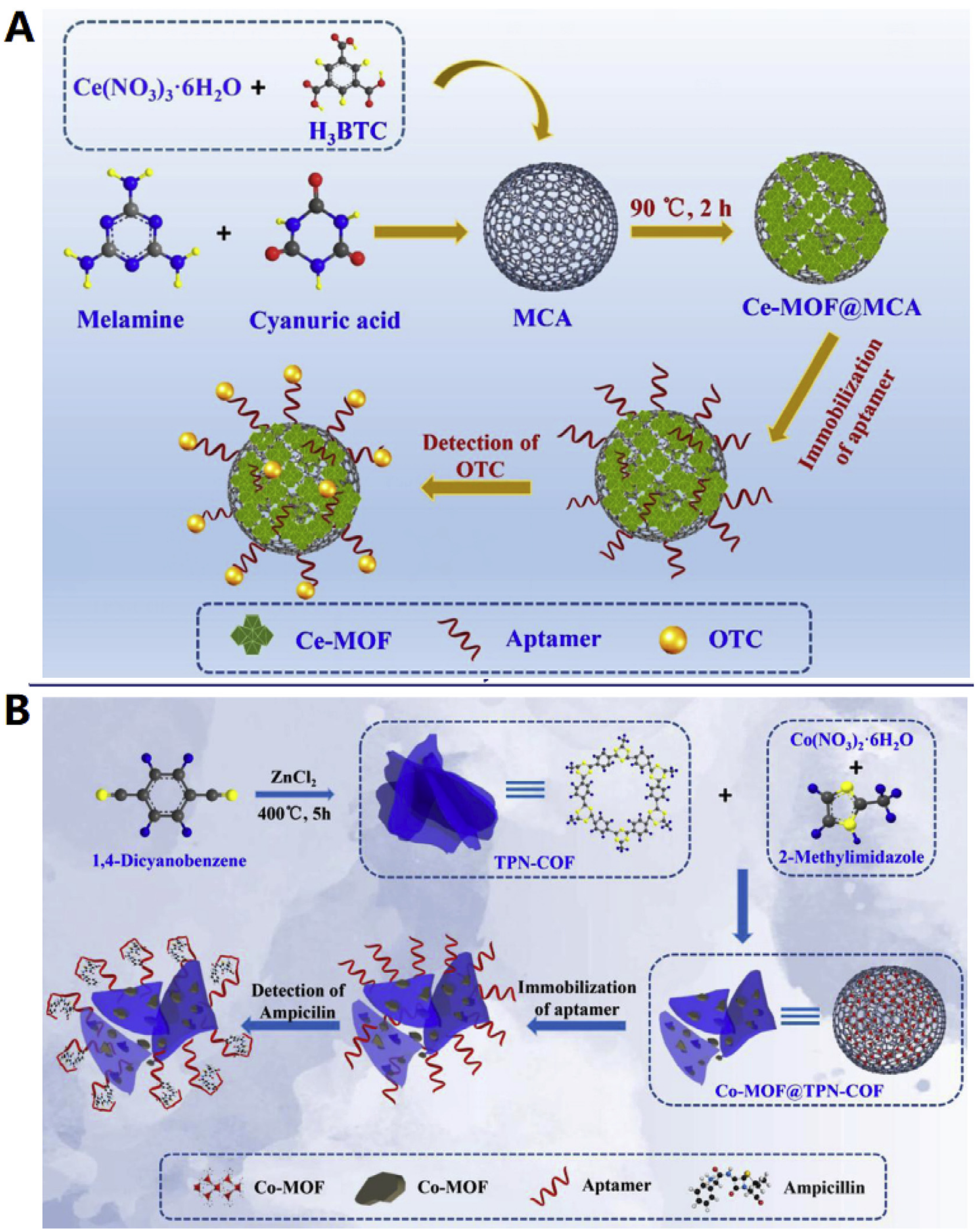

Fig. 4. (A) Schematic diagram of the fabrication procedure of the Ce-MOF@MCA-based aptasensor for detecting OTC(Zhou et al., 2019). (B) The schematic diagram of the construction of the Co-MOF@TPN–COF–based aptasensor for detecting ampicillin(Liu et al., 2019c).

chemical structure and crystal (e.g. individual frameworks, different Ce species of Ce-MOFs (Ce^{4+} and Ce^{3+}), and various functional amino-groups of COFs), and exhibited the interpenetrated morphologies and large specific surface area (Zhou et al., 2019). Similarly, Liu et al. synthesized a new nanoarchitecture of Co-MOF and terephthalonitrile (TPN)-based COF (Co-MOF@TPN-COF) as the label-free bioplatform for the detection of β-lactam antibiotic (Fig. 4B). The newly prepared porous hybrid materials (Co-MOF@TPN-COF) were fabricated through the addition of the synthesized TPN-COF into the preparation system of Co-MOF. The obtained Co-MOF@TPN-COF nanosheets showed an excellent electrochemical performance, highly specific surface area, and nitrogen-rich groups (Liu et al., 2019c). In addition, Sun and co-workers for the first time reported a novel palladium (Pd) doped core-shell MOFs@COFs (Pd/TiATA@LZU1) as a new multifunctional photocatalyst for photocatalysis. The Pd/TiATA@LZU1 with an excellent photocatalytic activity was prepared by using $-NH_2$ containing MOFs to directly grow the COF (2D iminie-based COF-LZU1) shell without additional functionalization (Sun et al., 2018a).

2.2.4. COFs-enzyme composites

Due to their high specific surface area, easily tunable pore size, and conveniently modified surface structure, COFs have been applied in enzyme immobilization to prepare COFs-enzyme composites (Wang et al., 2018a; Zhao et al., 2018). For example, Kandambeth et al. for the first time reported the COFs-enzyme composites, which was prepared through the immobilization of proteolytic enzymes into the COFs (Fig. 5A). In this study, through self-templated method, a hollow spherical COF (COF-DhaTab) with good chemical stability, mesoporous walls (3.7 nm), and large surface area (approximately 1500 m^2/g) was synthesized. Based on its unique mesoporous structure and great chemical stability, the prepared COF-DhaTab was used for the immobilization of trypsin to construct COF-DhaTab and trypsin composites (Kandambeth et al., 2015). Subsequently, Wang et al. rationally designed and successfully prepared a new COF-based trypsin (MG@TpPa-1-trypsin) for effectively and enzymatically digesting proteins. The MG@TpPa-1-trypsin was synthesized through the covalent immobilization of trypsin on the COFs-functionalized magnetic graphene by employing the glutaraldehyde as a coupling agent. The TpPa-1 was prepared by employing the paraphenylenediamine (Pa-1) and 1, 3, 5-triformylphloroglucinol (Tp) as the organic units. Benefiting from the merits of COFs and magnetic graphene, the obtained MG@TpPa-1-trypsin showed enhanced stability, good reusability, and improved digestion efficiency for proteins (Wang et al., 2017). In a recent study, Ma's group developed a novel biocomposites (lipase@COF–OMe) with the enhanced enzyme robustness. The use of monomers with specific functional groups could easily tune the pore environment of COFs to improve their compatibility with enzymes. By the designed interactions between COFs and enzymes, the enzyme active site orientation could be modulated for the improvement of biocomposite enzymatic performance. Moreover, for the first time, it was demonstrated that COFs with the tunable surface chemistry and unique mesoporous structure could provide a better microenvironment for the enhancement of enzymatic activity and higher affinity for the enzyme loading than other types of porous materials (e.g. MOFs and mesoporous silica) (Fig. 5B). In addition, the prepared lipase@COF–OMe possessed the advantages of greatly improved stability and easy recycling (Sun et al., 2018b).

2.2.5. Other COFs-based composites

Furthermore, there are some other COFs-based composites reported, such as COFs-quantum dots (QDs) composites, COFs-graphene

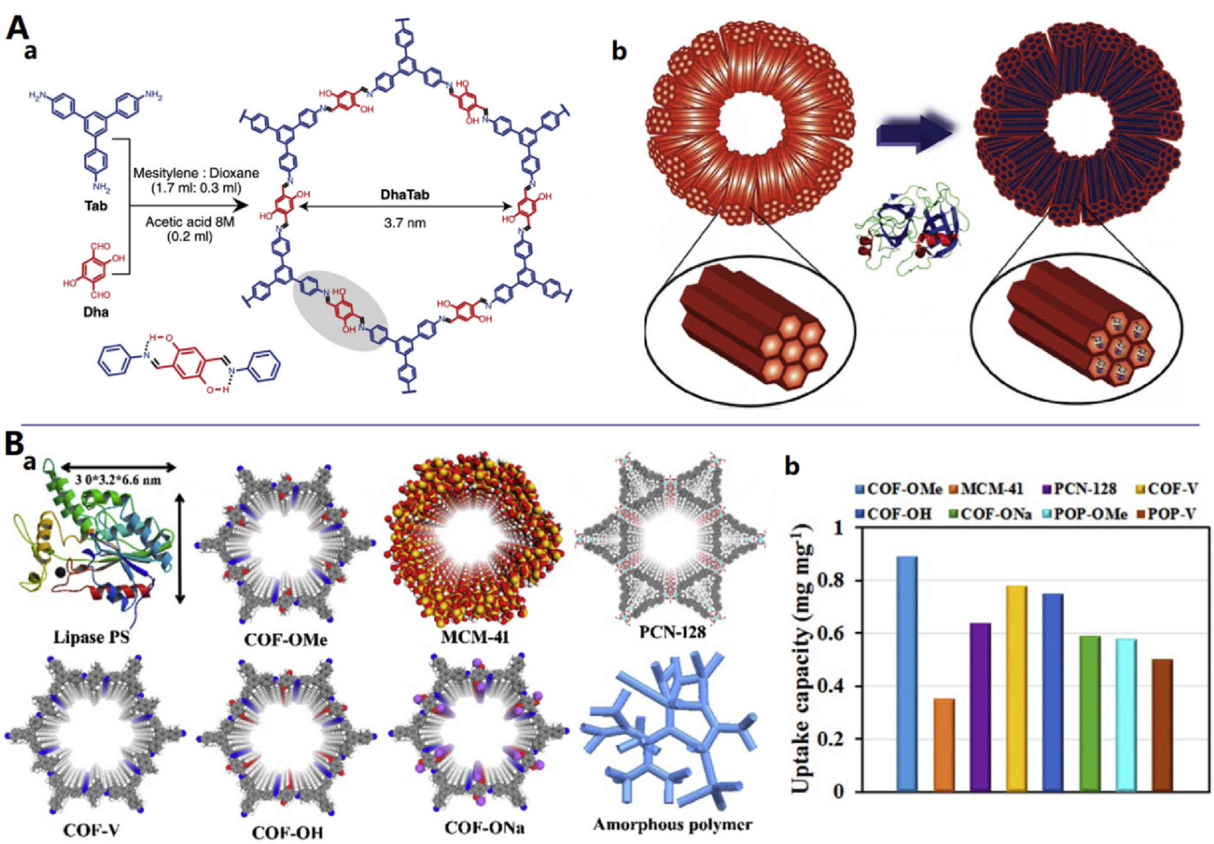

Fig. 5. (A) (a) Synthesis of COF-DhaTab by the Schiff base reaction of Tab and Dha. (b) Schematic representation of trypsin adsorption in COF-DhaTab(Kandambeth et al., 2015). (B) (a) Graphic view of lipase PS and porous materials used for the immobilization of enzymes (blue, N; gray, C; red, O; white, H; yellow, Si; purple, Na); (b) Enzyme uptake capacity of various porous materials after incubation in lipase PS solution (30 mg/mL) for 6 h (Sun et al., 2018b). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

composites, and so on. As reported by Liu's group, a new QDs-grafted COFs with thermal and chemical stability was prepared by the grafting of QDs on COFs through the Schiff base reactions between 3-aminopropyltriethoxysilane (APTEs)-modified CdSe/ZnS QDs and 1, 3, 5-triformylphloroglucinol-P-phenylenediamine (TpPa). The TpPa COFs as supports could be used to improve the sensitivity, and QDs modified via APTES could be used as a tentacle in sensitive and selective sensing of protein-bonding interactions (Ni et al., 2018). Soon after, their group developed 3D MIPs based on QDs-grafted COFs, which showed a strong optical response for quinoxaline-2-carboxylicacid (QCA) (Fig. 6A). In this study, the 1, 3, 5-triformylphloroglucinol-P-phenylenediamine as the COF material could be reacted with the amino-modified CdSe/ZnS QDs through Schiff-base reaction. Moreover, the as-prepared MIPs based on QDs-grafted COFs exhibited great thermal stability and chemical selectivity, achieving a highly selective and sensitive QCA detection (Zhang et al., 2019g). To simultaneously achieve functionalization and stabilization of absorbents, Wen et al. developed a novel graphene-synergized 2D COF (GS-COF) through the in situ loading of COFs (TDCOF) on the graphene sheets on the basis of a mutual promotion method (Fig. 6B). The oximation product *o*-TDCOF and *o*-GS-COF were synthesized, respectively. The results demonstrated that *o*-GS-COF had better acid and irradiation stability compared to that of the *o*-TDCOF. Moreover, compared to GO and *o*-TDCOF, the *o*-GS-COF possessed the stronger adsorption capacity for uranium. Furthermore, the *o*-GS-COF also showed the outstanding selectivity for uranium (Wen et al., 2018). In addition, to achieve a selective and specific sensing, the aptamers are attached on the COFs by π-π stacking interaction due to their rich functional groups and π-conjugation frameworks (Wang et al., 2019; Yan et al., 2019).

2.3. Molecularly imprinted COFs

Molecularly imprinted polymers (MIPs) as a biomimetic recognition material has attracted much attention due to its wide applications in the

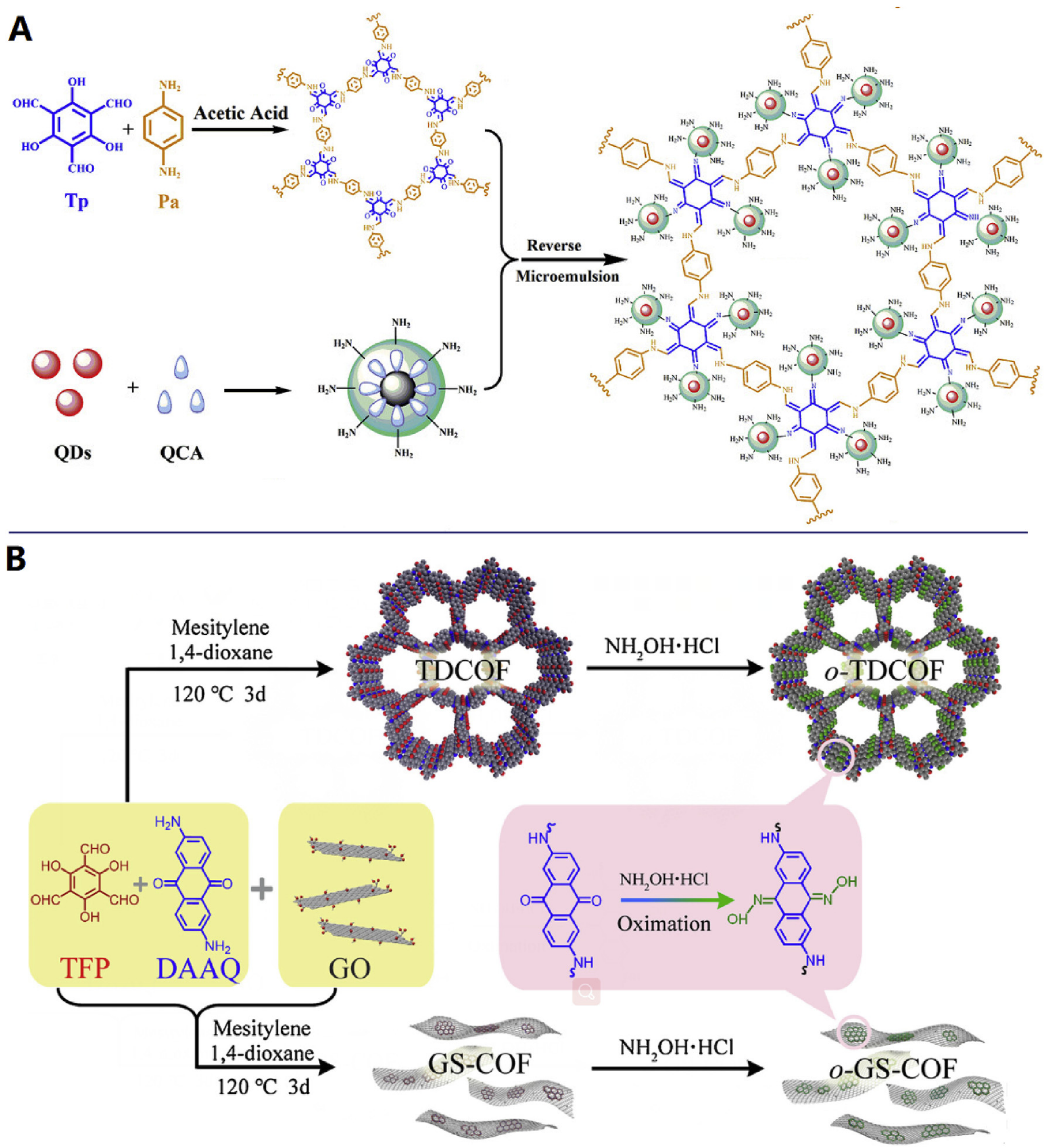

Fig. 6. (A) Scheme for the preparation of quantum dots (QDs)-grafted COFs (Zhang et al., 2019g). (B) Preparation of TDCOF, GS-COF, *o*-TDCOF and *o*-GS-COF (Wen et al., 2018).

improvement of the sensitivity and selectivity of analytical methods by selectively capturing the structurally related groups or small organic molecules (Jiang et al., 2007). Based on their merits of low cost, highly mechanical and chemical stability, outstanding recognition properties, easy preparation, the molecularly imprinted strategy has been used to prepare the molecularly imprinted COFs (MICOFs) for the improvement of COF performances (Liu et al., 2019a; Zhang et al., 2019b; Zhang et al., 2019g). As Ji's group reported, the imine-linked MICOFs were for the first time synthesized by a general and efficient synthetic approach. By utilizing fenvalerate as a template, the Schiff base was rapidly formed through the reaction between 1, 3, 5-triformylphloroglucinol (Tp) and 1, 3, 5-tris (4-aminophenyl) benzene (TAPB) under the room temperature condition with the existence of $Sc(OTf)_3$. The as-prepared MICOFs showed an excellent selectivity for four cyano pyrethroids with similar structure (Ji et al., 2018a). It was worth noting that Zhang et al. successfully synthesized a novel dual-function molecularly-imprinted optopolymer (MIOP) on the basis of QDs-grafted COFs. In this study, the QDs modified by 3-aminopropyltriethoxysilane and tetraethoxysilane could provide the $-NH_2$ surface binding sites for the covalent interaction with TpPa COFs through the Schiff base reaction. The QDs' partial amino groups achieved a non-covalent interaction with tyramine (TYM). The as-obtained MIOP based on QDs-grafted COFs as an excellent sorbent could detect tyramine via the optosensing and SPE-HPLC (Zhang et al., 2019a). Li et al. prepared a new nanocomposite consisting of a MICOFs shell and an amorphous seed through a heterogeneous nucleation growth strategy (Fig. 7A). By employing the ibuprofen as a dummy template, the novel MICOFs with the large surface areas were synthesized via 4, 4′-diaminobiphenyl and 1, 3, 5-triformylbenzene, and then were placed on the surfaces of monodisperse amorphous seeds. Due to the strong π-interaction, the as-synthesized MICOF@SiO_2 nanocomposite showed excellent adsorption abilities, good selectivity, and binding kinetics for the nonsteroidal anti-inflammatory drugs (NSAIDs) (Li et al., 2019b). In a very recent study, by one-pot reverse microemulsion polymerization, Liu et al. successfully prepared a MIP doped with a COF grafted onto carbon nanodots (CNs) to construct fluorescent nanoprobe for the detection of 4-ethylguaiacol (4-EG) (Fig. 7B) (Liu et al., 2019a).

3. Applications of functionalized COFs to sensing

Based on their diverse compositions and synergistic functions, the functionalized COFs are endowed with unique sensing performances, and have been successfully applied in sensing, such as colorimetric sensing, fluorescent sensing, and electrochemical sensing, and QCM sensing (Chen et al., 2019b; He et al., 2019b; Wu and Yang, 2017). In a previous review, we have systematically summarized applications of COFs in sample pretreatment (Li et al., 2018a). Herein, the applications of functionalized COFs to sensing are comprehensively summarized (Table 1).

3.1. Colorimetric sensing

Due to its easy readout and rapid visual assay via the naked eyes or low-cost and portable device, colorimetric sensing has received much

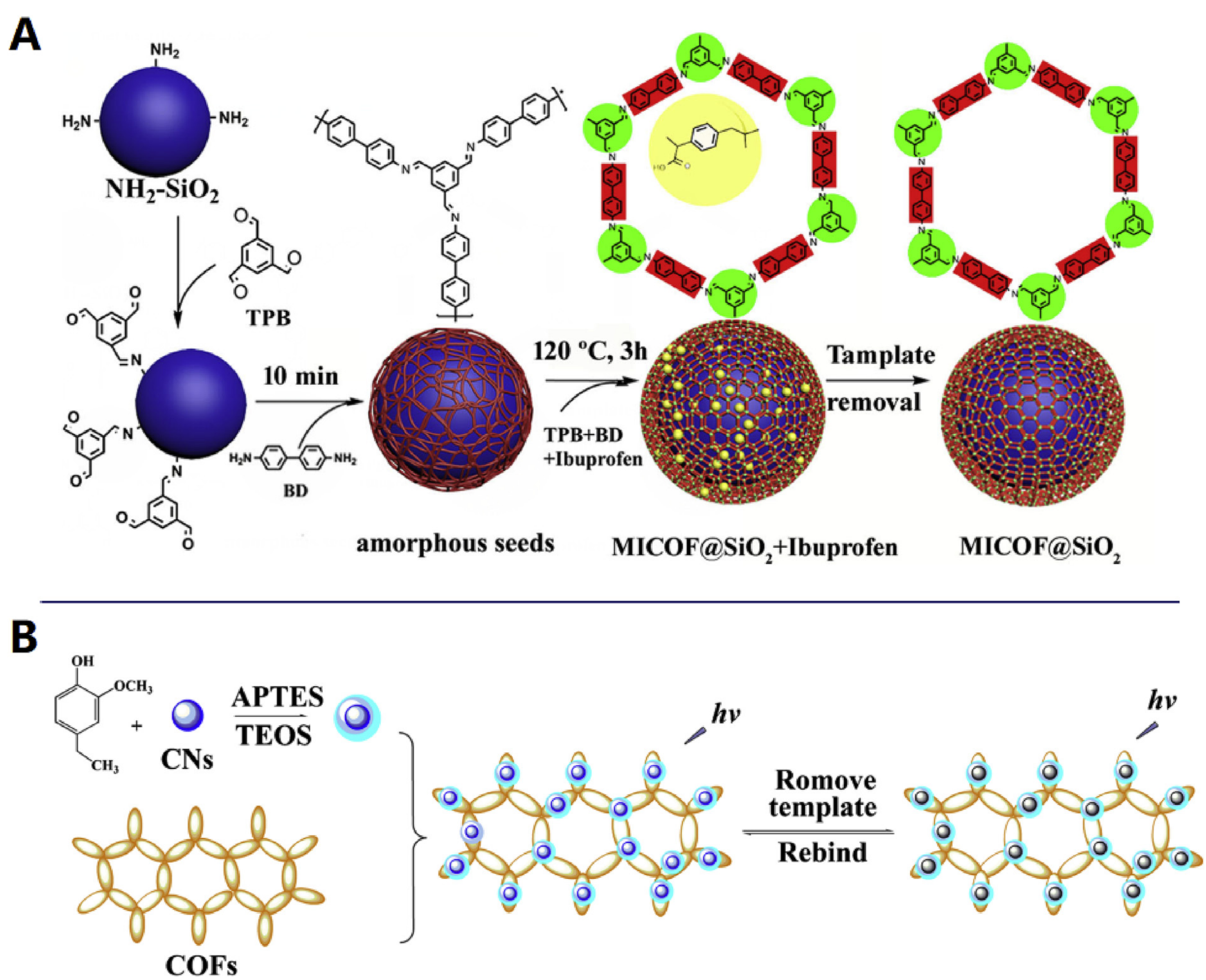

Fig. 7. (A) The synthesis route for MICOF@SiO_2 nanocomposite (Li et al., 2019b). (B) A schematic representation of the synthesis of CN-grafted COF@MIP by a one-pot room temperature synthesis strategy with reverse microemulsion polymerization (Liu et al., 2019a).

Table 1
Applications of reported functionalized covalent organic frameworks to sensing.

Sensing modes	Functionalized COFs	Target analytes	Linear range	LOD	Ref.
Colorimetric sensing	COF-HQ	pH	1–5	/	Chen et al. (2018)
	Fe–COF	H_2O_2	7–500 μM	1.1 μM	Wang et al. (2018b)
	Fe–COF	Glucose	5–350 μM	1.0 μM	Wang et al. (2018b)
	AuNPs@Tp-Bpy	Hg^{2+}	/	0.33 nM	Cui et al. (2019b)
Fluorescent sensing	COF-HQ	pH	1–5	/	Chen et al. (2018)
	SNW-1	TNT	/	0.51 ppm in solution; 9.8 ppb in vapor	Zhang et al. (2012)
	QD-grafted COFs	Protein	/	5.4×10^{-4} mg/mL	Ni et al. (2018)
	HHTP-DPB COF	Formaldehyde	/	/	Wang et al. (2018c)
	QDs-grafted COFs	Tyramine	$35–3.5 \times 10^4$ μg/kg	7.0 μg/kg	Zhang et al. (2019a)
	CN-embedded COFs	Tryptamine	0.025–0.4 mg/kg	7.0 μg/kg	Zhang et al. (2019b)
	CN-grafted COFs@MIPs	4-ethylguaiacol	0.025–1 μg/mL	17 ng/mL	Liu et al. (2019a)
	MIPs Based on QDs-Grafted COFs	Quinoxaline-2-carboxylicacid	1–50 μmol/L	0.85 μmol/L	(Zhang et al., 2019g)
	COF-LZU8	Hg^{2+}	/	/	Ding et al. (2016)
	PI-COF	TNP	0.5–10 μM	0.25 μM	Zhang et al. (2017)
	TFPPy-DETHz-COF	Fluoride anion	/	At a ppb level	Li et al. (2018e)
	EB-TFP-iCONs	DsDNA	/	/	Mal et al. (2018)
	Bth-Dma COF	Fe^{3+}	/	/	Chen et al. (2019a)
Electrochemical sensing	Pt-COFs	C-reactive protein	1–400 ng/mL	0.2 ng/mL	Liu et al. (2016)
	TAPB-DMTP-COF	Lead	0.005–2.0 μmol/L	1.9 nmol/L	Zhang et al. (2018d)
	TAPB-DMTP-COFs/AuNPs	Chlorogenic acid	$1.0 \times 10^{-8}–4.0 \times 10^{-5}$ mol/L	9.5×10^{-9} mol/L	Zhang et al. (2018c)
	Au-COFs	Cardiac troponin I	0.5 pg/mL-10 ng/mL	0.17 pg/mL	Zhang et al. (2018e)
	Co-MOF@TPN-COF	Ampicillin	1.0 fg/mL-2.0 ng/mL	0.217 fg/mL	Liu et al. (2019c)
	NH_2-MWCNT@COF	Sulfamerazine	$3.0 \times 10^{-7}–2.0 \times 10^{-4}$ mol/L	1.1×10^{-7} mol/L	Sun et al. (2019)
	p-COF	Epidermal growth factor receptor	0.05–100 pg/mL	5.64 fg/mL	Yan et al. (2019)
	p-COF	Living cancer cell	500×10^5 cell/mL	61 cell/mL	Yan et al. (2019)
	Py-M-COF	Enrofloxacin	/	6.07 fg/mL	Wang et al. (2019)
	Py-M-COF	Ampicillin	/	0.04 fg/mL	Wang et al. (2019)
QCM sensing	COFs-AuNPs	Aflatoxin B1	0.05–75 ng/mL	2.8 pg/mL	Gu et al. (2019)
	BABE-TFPy COF	2-chloroethyl ethyl sulfide	5.6–19.7 ppm	0.96 ppm	He et al. (2019b)
Photoelectrochemical sensing	p-COFs	C-reactive protein	0.5–100 ng/mL	0.1 ng/mL	(Zhang et al., 2019e)
Humidity sensing	COF-TXDBA	Water molecules	/	/	(Singh et al., 2017b)
Enantioselective sensing	BINOL-based COFs	Chiral odor vapors	/	/	Wu et al. (2019)

Note:/, not reported.

attention and been widely applied in detection of the analytes on the basis of the color change(Kim et al., 2012; Weerathunge et al., 2019). Recently, some functionalized COFs with enzyme-like catalytic activity have been developed to catalyze chromogenic substrate such as 3, 3′, 5, 5′-tetramethylbenzidine (TMB) for the generation and amplification of colorimetric signal. As shown in Fig. 8A, Wang et al. for the first time developed a novel Fe-porphyrin-based COF (Fe–COF) with an excellent peroxidase-like activity by a facile post-synthetic method. The as-prepared Fe–COF showed the merits of good stability, ultrahigh catalytic efficiency, and easy preparation. With the existence of H_2O_2, the prepared Fe-COFs could be used to catalyze the chromogenic substrate TMB to generate a blue color. Surprisingly, compared to the biological enzyme horseradish peroxidase (HRP), the prepared Fe-COFs possessed a higher affinity toward the TMB and H_2O_2. On this basis, the prepared Fe-COFs were used as the peroxidase mimic for construction of colorimetric sensor. With this colorimetric sensor, the H_2O_2 could be sensitively detected ranging from 7 to 500 μM, and with a low limit of detection (LOD) of 1.1 μM. Furthermore, the Fe–COF combined with glucose oxidase (GOx) could achieve a sensitively quantitative detection of glucose in human serum samples through a one-pot colorimetric strategy. The concentration of glucose could be determined with a detection range of 5–350 μM, and with a low LOD of 1.0 μM (Wang et al., 2018b). Recently, Cui et al. innovatively synthesized bipyridine-containing COF nanosheets (Tp-Bpy NSs) with the abundant nitrogen-containing functional groups and regular pore structures, and then abundant nitrogen-containing functional groups of Tp-Bpy NSs were employed as active sites for the in situ production of AuNPs on the Tp-Bpy NSs to generate AuNPs@Tp-Bpy nanocomposite. The AuNPs were anchored onto the Tp-Bpy NSs by the coordination bonds, endowing the AuNPs with strong stability, good dispersibility, and high catalytic performance. Moreover, the anchored AuNPs on the Tp-Bpy NSs were fully exposed for the enhancement of their peroxidase-like activity based on the 2D regular pore structure of Tp-Bpy NSs. With the existence of Hg^{2+}, the as-obtained AuNPs@Tp-Bpy nanocomposite with high stability and outstanding water dispersibility could catalyze the oxidation of chromogenic substrate TMB to produce a significant blue color in the presence of H_2O_2 due to the Hg^{2+}-triggered enhanced peroxidase-like activity. On this basis, a sensitive and high-performance colorimetric sensor was developed for the determination of Hg^{2+} in aqueous solution (Fig. 8B). The Hg^{2+} in environmental samples could be stably and sensitively determined with an ultralow LOD of 0.33 nM (Cui et al., 2019b). In addition, by utilizing 8-hydroxyquinoline-functionalized COF (COF-HQ), Wang et al. developed a novel dual-mode colorimetric and fluorescent pH sensor for accurate and real-time determination of pH in aqueous solution. In the colorimetric mode, COF-HQ exhibited a significant color change from yellow to black with the increase of acidity, which could be employed as a colorimetric sensor for the determination of pH. Furthermore, the COF-HQ emission

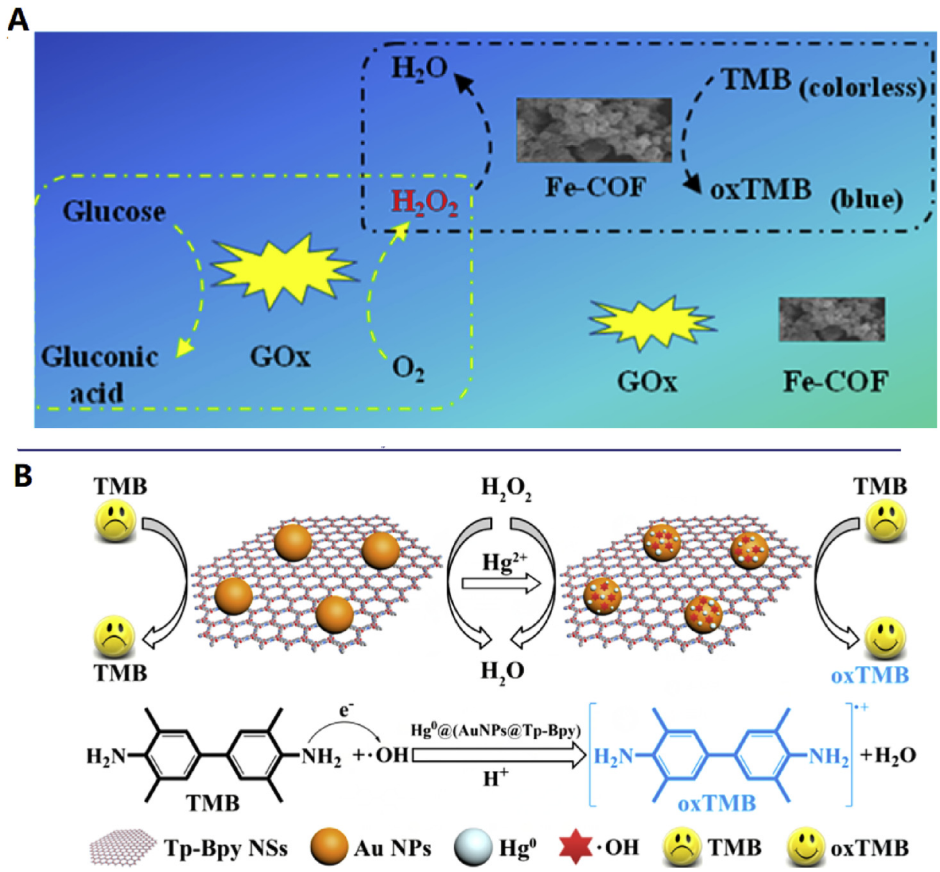

Fig. 8. (A) Schematic representation of a colorimetric sensor for glucose detection using the Fe–COF as the catalyst(Wang et al., 2018b). (B) Mechanism of colorimetric detection of mercury ions using AuNPs@Tp-Bpy(Cui et al., 2019b).

intensity weakened with the decrease of pH, which was used as fluorescent pH sensor and showed a great linear relationship for pH ranging from 1 to 5 (Chen et al., 2018). Although colorimetric sensing methods have exhibited some merits (e.g. easy readout, rapid visual assay, and without requirement of equipment), their accuracy and sensitivity maybe easily influenced by the interferences generated from the sample background color. In addition, the colorimetric sensing methods are still inferior to the fluorescence sensing methods in respects of sensitivity and anti-interference (Tang et al. 2017, 2019).

3.2. Fluorescent sensing

Fluorescence sensing is established mainly on the basis of the target analytes mediated fluorescence quenching (“turn-off”) or fluorescence enhancement (“turn-on”) (Li et al. 2016a, 2019c; Zhou et al., 2014). Owing to its advantages of portability, operational simplicity, excellent sensitivity, real-time assay, and outstanding selectivity, fluorescence sensing has attracted much attention and been widely applied in multiple fields (Li et al., 2016a; Liu et al., 2018; Lu et al., 2016; Tian et al., 2017). Compared with colorimetric sensing, fluorescence sensing often features with higher capacity of anti-inference, high sensitivity, and lower requirements of sample amounts (Tang et al., 2019). In recent years, some functionalized COFs with fluorescence performances have been developed for the construction of fluorescent sensors (Li et al., 2017; Ni et al., 2018; Zhang et al., 2019b). For example, Ding et al. synthesized a novel functionalized COF (thioether-based fluorescent COF, named COF-LZU8) under solvothermal conditions for fluorescence sensing and facile removal of Hg^{2+} (Fig. 9A). In this study, the π-conjugated framework of COF-LZU8 served as a signal transducer, and the densely distributed thioether groups were used as the receptor for Hg^{2+}, and the regular pores were advantageous for the real-time determination and mass transfer. The COF-LZU8 with outstanding performances of excellent selectivity, high sensitivity, and real-time response, and the easy visibility could achieve an excellent fluorescence sensing. Moreover, COF-LZU8 was used for the efficient removal of Hg^{2+} from water (Ding et al., 2016). Zhang et al. synthesized a novel fluorescent polyimide COF (PI–COF) via solvothermal strategy utilizing perylenetracarboxylic dianhydride and tetra (4-aminophenyl) porphyrin, which showed excellent thermal stability and porous crystalline. By a facile liquid phase exfoliation method, the few-layered highly fluorescent PI covalent organic nanosheets (PI-CONs) could be prepared from the fluorescent PI-COFs. The as-obtained PI-CONs were used as an efficient fluorescent probe for the selective and sensitive determination of 2, 4, 6-trinitrophenol (TNP)due to the combination between inner filter effect (IFE) and electron transfer (ET) (Fig. 9B). The developed fluorescence sensing method could detect TNP with a good linear response ranging from 0.5 to 10 μM, and with a low LOD of 0.25 μM (Zhang et al., 2017). Utilizing self-exfoliates of ethidium bromide (EB) based COFs (EB-TFP) in water, Ajayaghosh and co-workers developed the novel 2D ionic covalent organic nanosheets (EB-TFP-iCONs) for sensitively and selectively detecting double-stranded DNA (dsDNA). With the existence of dsDNA, the self-exfoliated EB-TFP-iCONs in aqueous medium would reassemble, leading to the generation of new hybrid EB-TFP-iCONs-DNA crystalline nanosheets with an enhanced fluorescence at 600 nm. On this basis, the complementary DNA strands could be sensitively detected by employing the prepared EB-TFP-iCONs as the 2D fluorescent platform (Mal et al., 2018). Through a pinpoint surgery on the pore walls, Li et al. converted less emissive COFs into light-emitting materials. The ET from the linkage to COF was eliminated due to the deprotonation of N^- anion by scissoring the N–H unit, and the fluorescence quenching was suppressed, leading to the enhancement of light-emitting performance in a proportional fashion. The

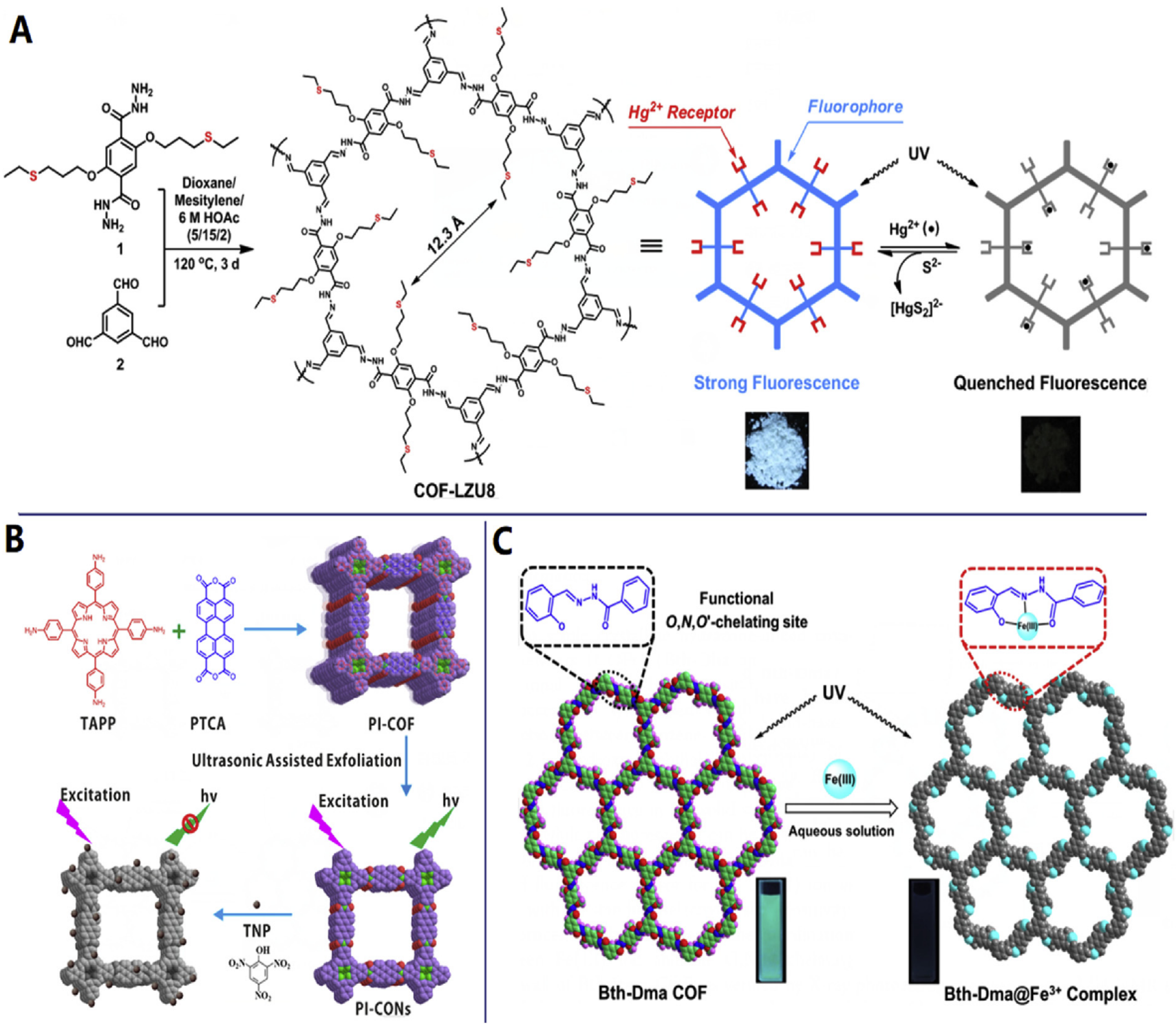

Fig. 9. (A) Synthesis of COF-LZU8 and its applications in both fluorescence detection and removal of Hg^{2+} from water(Ding et al., 2016). (B) Schematic representation for the preparation of the fluorescent PI-CONs and their application in the determination of TNP(Zhang et al., 2017). (C) Construction of proposed fluorescence sensor for highly selective and sensitive metal ion detection(Chen et al., 2019a).

F^- anion could trigger the improvement of light-emitting activity, while NO_3^- and other halogen anions were inert. On this basis, F^- could be detected by the proposed fluorescence switch-on sensing renders at a ppb level (Li et al., 2018e). In a very recent study, Chen et al. rationally designed and successfully fabricated two stable O, N, O'-chelating sites functionalized crystalline hydrazone-linked COFs (Bth-Dma and Bth-Dha) through the Schiff-base condensation reactions between the benzene-1, 3, 5-tricarbohydra-zide (Bth) and 2, 5-dimethoxyterephthal-aldehyde (Dma) or 2, 5-dihydroxyterephthalaldehyde (Dha), respectively. Interestingly, Bth-Dma showed a strong fluorescence in an aqueous dispersion and in the solid state, while Bth-Dha exhibited no fluorescence. Based on the coordination interaction between the Fe (III) ion (Fe^{3+}) and the O, N, O'-chelating sites of Bth-Dma COF pore wall, the as-prepared Bth-Dma could be utilized as a turn-off fluorescence sensor for sensitive and selective detection of Fe^{3+} ions in water solution(Fig. 9C)(Chen et al., 2019a). Currently, most of COF-based fluorescence sensors are proposed based on the "turn-off" mode (the quenching effect of targets on COF fluorescence) for detection of explosives/explosive-like molecules and heavy metal ions (Liu et al., 2019d). COF-based fluorescence sensors for the determination of Hg^{2+} may be attributed to the combination of Hg^{2+} to the S atoms, resulting in the transfer of electrons from the π-conjugation framework to the unoccupied orbitals of Hg^{2+}(Ding et al., 2016). However, other metal ions (e.g. Co^{2+}, Pb^{2+}, Mg^{2+}, Ca^{2+}, Cu^{2+}, Mn^{2+}, Fe^{3+}, Al^{3+}, and Au^{+}) also may cause fluorescence quenching of functionalized fluorescence COFs, which limited the detection selectivity in a certain degree (Chen et al., 2019a; Li et al., 2016c; Liu et al., 2019d; Xue et al., 2017). Therefore, the development of novel COFs modifying by different functional groups for the improvement of fluorescence sensor selectivity is extremely promising. Moreover, functionalized fluorescence COFs remain to be developed for sensing many other interesting targets, and COF-based fluorescence sensors based on the "turn-on" mode also need be further studied (Liu et al., 2019d). In addition, COF with stronger fluorescence performance should be developed for sensor construction (Xue et al., 2017).

3.3. *Electrochemical sensing*

Electrochemical sensing is mainly established on the basis of the changes of output-electrical signals generated through the chemical reactions of target analytes with the electrode-immobilized recognition elements. The production of electrical signals is related to the target analyte concentrations, achieving a qualitative determination and a quantitative detection of target molecules (Wang et al., 2019; Zhang et al., 2018e; Zhang et al., 2019f). Electrochemical sensing recently has caused wide attention in multiple fields, such as environmental monitoring(Urbanová et al., 2017), clinical diagnosis(Johari-Ahar et al., 2018), food safety(Lv et al., 2018), and pharmacy(Kubendhiran et al., 2018) owing to its sensitive response, low cost, and simple operation(Liu et al., 2019e). Due to their intrinsically low electrical conductivity, COFs are rarely employed for the constructions of electrochemical sensors. Fortunately, some functionalized COFs (e.g. COFs@Pt NPs, COFs@Au NPs, COFs@MOFs, and so on) have been successfully developed for electrochemical sensing (DeBlase et al., 2015; Liu et al., 2016; Sun et al., 2019; Zhang et al. 2018c, 2018d, 2018e). As shown in Fig. 10A, Zhang et al. developed a new sensitive sandwich-type electrochemical immunosensor for the quantitative determination of cardiac troponin I (cTnI) through a signal amplification method that employed the AuNPs doped COFs and electron mediator toluidine blue (TB-Au-COFs) as labels for the amplification of electrochemical signal. The proposed electrochemical sensor showed acceptable reproducibility, good accuracy, and high stability, and could be used to accurately and rapidly detect the cTnI with a linear range of 0.5 pg/mL-10.0 ng/mL, and with a low LOD of 0.17 pg/mL (Zhang et al., 2018e). Liu et al. synthesized a novel porous hybrid material of Co-MOF@TPN-COF with an outstanding electrochemical activity, nitrogen-rich groups, and high specific surface area as a bioplatform for the construction of electrochemical sensor. The developed electrochemical sensor could achieve an ultrasensitive detection of ampicillin (AMP) with a low LOD of 0.217 fg/mL within the concentration of AMP ranging from 1.0 fg/mL to 2.0 ng/mL (Liu et al., 2019c). Sun et al. developed a new 3D electrochemical sensor with selective recognition and excellent signal amplification of sulfamerazine (SMR) based on the glassy carbon electrode (GCE) modified by the MoS_2 nanosheets and amino-functionalized carbon nanotubes@COFs (NH_2-MWCNT@COF) with high conductivity. To selectively recognize the SMR, MIP membranes were anchored on the modified GCE surface through an electrochemical polymerization. The as-obtained sensor showed an excellent selectivity for the SMR and a good reproducibility, and accomplished a sensitive determination of SMR with a low LOD of 1.1×10^{-7} mol/L. Moreover,

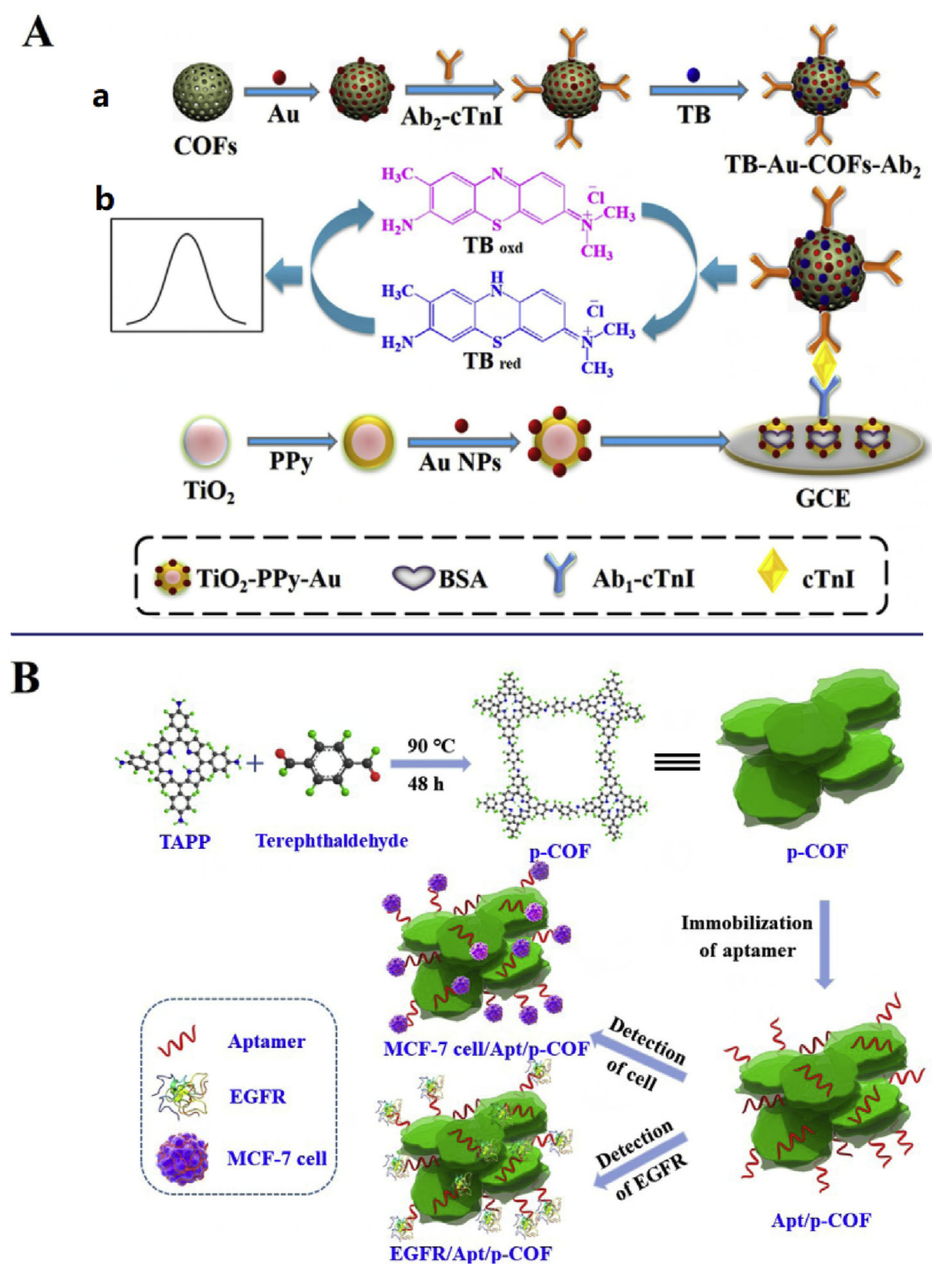

Fig. 10. (A) Schematic representation for the preparation procedure of the TB-Au-COFs-Ab_2 labels and the schematic illustration of the sandwich-type electrochemical immunosensor(Zhang et al., 2018e). (B) Schematic diagram of the p–COF–based aptasensor for detecting the EGFR or MCF-7 cells (Yan et al., 2019).

the proposed sensor was successfully used for the detection of SMR in chicken and pork samples with good recoveries of 86.0–102.0% (Sun et al., 2019). By utilizing a simple oil-bath strategy, Yan et al. for the first time prepared a novel porphyrin-based COF (p-COF) as a new sensing layer to immobilize the epidermal growth factor receptor (EGFR)-targeting aptamer strands for the determination of trace EGFR and the living michigan cancer foundation-7 (MCF-7) cells (Fig. 10B). The as-synthesized p-COF (with a nanosheet-like structure, rich nitrogen-bearing groups, and large cavities) exhibited excellent electrochemical activity, high stability, good bioaffinity, and low toxicity in water solution. Owing to the production of an aptamer-EGFR complex through interactions of the aptamer strands with EGFR, the electrochemical signals were significantly changed. On this basis, the electrochemical sensor with good stability, high selectivity, favorable applicability, and acceptable recyclability was constructed for the determination of trace EGFR and living MCF-7 cells. With this electrochemical sensor, the EGFR could be detected with a wide linear determination range from 0.05 to 100 pg/mL, showing an extremely low LOD of 5.64 fg/mL. For living MCF-7 cells, the proposed electrochemical sensor showed a low LOD of 61 cell/mL (Yan et al., 2019). Based on the condensation polymerization between melamine and 1, 3, 6, 8-tetrakis (4-formylphenyl) pyrene through imine bonds, Wang et al. developed a new COF (Py-M-COF) with large specific surface area, nanosheet-like structure, extended π-conjugation framework, big pore cavities, and rich functional groups (e.g. C═O, C═N, and C═C) to establish an electrochemical aptasensor for the ultrasensitive assay of

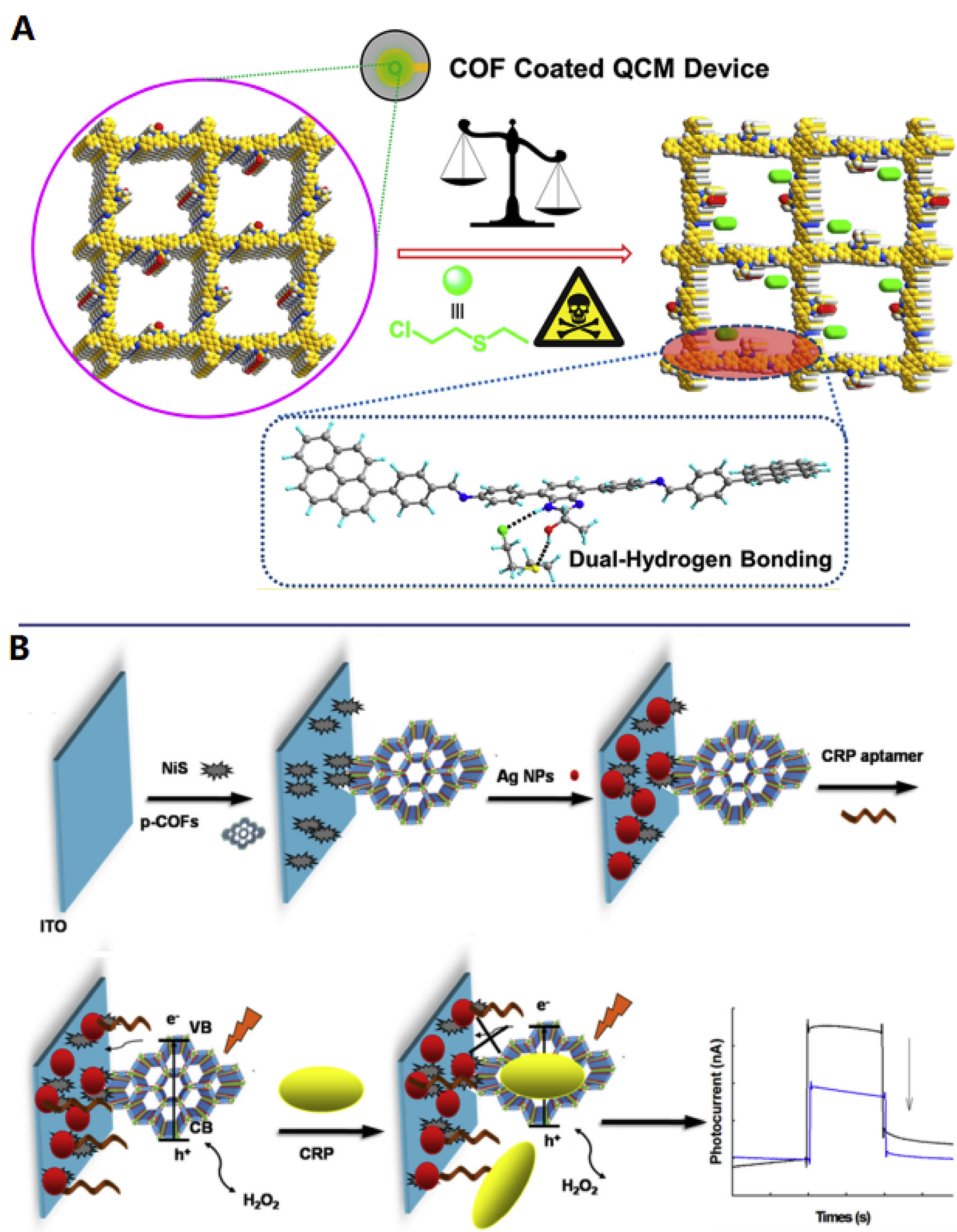

Fig. 11. (A) Schematic diagram of the benzimidazole-containing COF-based QCM sensor for exceptional detection of CEES(He et al., 2019b). (B) Schematic representation of procedure for the stepwise fabrication of CRP photoelectrochemical biosensors (Zhang et al., 2019e).

antibiotics(including ampicillin (AMP) and enrofloxacin (ENR)). With this electrochemical aptasensor, the AMP and ENR could be detected with low LODs of 0.04 and 6.07 fg/mL, respectively(Wang et al., 2019). Currently, electrochemical sensing has been widely developed and applied in many fields, but it still suffers from the poor repeatability, low stability, and the requirement of analytes' outstanding electroactivity. To overcome these defects and improve its accuracy and stability, a novel type of ratiometric electrochemical sensing has been proposed, which shows great prospects (Chai et al., 2013).

3.4. Other sensing

In addition, some other sensors based on the functionalized COFs have been reported, such as QCM sensor(He et al., 2019b), photoelectrochemical aptasensor(Zhang et al., 2019e), humidity aptasensor (Singh et al., 2017a), and so on(Wu et al., 2019; Zhu et al., 2010). As Fig. 11A showed, He and co-workers for the first time developed a novel COF coated QCM sensor for the determination of multiple volatile organic compounds (VOCs) based on a novel benzimidazole-containing COF (BABE-TFPy COF) with the advantages of excellent chemical stability, high porosity, abundant accessible benzimidazole sites, and crystallinity synthesized through the Schiff base condensation between the 1,3,6,8-tetrakis(4-formylphenyl)pyrene (TFPy) and 1-(4,7-bis(4-aminophenyl)-1H-benzoimidazole-2-yl)ethan-1-ol(BABE). Compared to other common VOCs, the developed BABE-TFPy COF coated QCM sensor showed a higher sensitivity for the detection of 2-chloroethyl ethyl sulfide (CEES). In addition, the frequency variation of the prepared COF coated QCM sensor was linearly related to the concentrations of CEES vapor ranging from 5.6 to 19.7 ppm, and with an extremely low LOD of 0.96 ppm (He et al., 2019b). In another study, Wang and co-workers innovatively developed a new QCM sensor based on the AuNPs doped molecularly imprinted layer and COFs-AuNP composite for the determination of aflatoxin B1. The QCM sensor showed high sensitivity and good selectivity due to the loading capacity of the COFs-AuNPs composites and the specific recognition effect of MIP of poly (*o*-ATP) with AuNPs. The proposed QCM sensor could detect aflatoxin B1 with a wide linear range of 0.05–75ng/mL, showing a low LOD of 2.8 pg/mL (Gu et al., 2019). On the basis of 2D porphyrinic COFs (p-COFs), Zhang et al. developed a new photoelectrochemical (PEC) aptasensor for the non-label determination of C-reactive protein (CRP) (Fig. 11B). In this study, the p-COFs were synthesized by Schiff base condensation and showed high conductivity. Owing to strong and rigid covalent linkages, the stability of p-COFs was significantly improved. Meanwhile, the photocurrent conversion efficiency was improved due to the introduction of p-COFs hindering the recombination between the electrons and holes. And the prepared p-COFs showed the enhanced photocurrent intensity compared to pure porphyrin. Because of the photo-generation of holes on light excitation, the signal was amplified through the oxidation of the electron donor H_2O_2. The aptamer specifically recognized via the CRP was assembled on the surface of AgNPs. The introduction of CRP hindered the ET, which leaded to the photocurrent response decrease. On this basis, PEC aptasensor with high stability, rapid response, excellent selectivity and wide linear range was successfully constructed for the detection of CRP. The CRP could be detected with a wide linear range of 0.5–100 ng/mL and a low LOD of 0.1 ng/mL (Zhang et al., 2019e). In addition, Singh et al. synthesized a new Truxene based porous, crystalline COFs (COF-TXDBA) by employing the Truxene as a building block for the construction of humidity sensing. The proposed humidity sensing showed a good response owing to the existence of boronic ester linkages and the interaction between boronic ester linkages and water molecules on the planar sheets of COF backbone. Attributed to the ordered micropores/mesopores and high surface area in this COF-TXDBA, plenty of boronic ester active sites were exposed to interact with water molecules (Singh et al., 2017b).

4. Conclusion and prospective

COFs as an emerging class of microporous organic polymers have been extensively developed and applied in multiple fields. Recently, to improve the potential performances of COFs and extend their applicability, a great many COFs with different functionalities have been widely explored through functional modification, and have received increasing concerns in the fields of sensing. In this review, firstly, the design and construction of functionalized COFs is comprehensively summarized, including COFs with post-synthetic modification, COFs-based composites (e.g. COFs-metal nanoparticle composites, COFs-metal oxide nanoparticle composites, COFs-MOFs composites, and COFs-enzyme composites), and MICOFs. Then, the attentions are concentrated on the applications of functionalized COFs in the fields of sensing, including colorimetric sensing, fluorescent sensing, electrochemical sensing, and other sensing. To promote the development on construction and applications of functionalized COFs, the following challenges and opportunities should be considered in future work:

(1) Post-synthetic modification strategies have played a key role in the development and construction of functionalized COFs, which can endow COFs with some distinctive functions to greatly broaden their potential in future applications. However, the post-synthetic modification of COFs is mainly limited to the channel wall functionalization, suffering from some extra obstacles in precise control of functional group amount and distribution within COFs. Therefore, the COFs with post-synthetic modification remain to be further studied to improve potential performances of COFs.
(2) To date, plenty of COFs-based composites have been successfully prepared through integration of functional materials (e.g. MNPs, metal oxide NPs, MOFs, biological enzymes, and QDs) with COFs for improving their performances and broadening their applications. However, among these reported COFs-based composites, COFs-enzyme composites are rarely reported and they are prepared by the immobilization of only an enzyme on the COFs (Kandambeth et al., 2015; Sun et al., 2018b; Wang et al., 2017). Based on their high specific surface area, easily tunable pore size, and conveniently modified surface structure, COFs may be further explored for the immobilization of multiple enzymes to prepare novel COFs-enzymes multifunctional composites for sensing and biocatalytic cascades.
(3) Although some functionalized COFs have been used to improve the sensitivity, reproducibility, and stability in sensing applications, sensing modes are relatively single. Therefore, applications of functionalized COFs in sensing remain to be further investigated. For example, some functionalized COFs can be developed for construction of new sensing strategies, such as chemiluminescent sensing, surface-enhanced Raman scattering (SERS) sensing, surface plasmon resonances (SPRs) sensing, and electrochemiluminescence sensing.

CRediT authorship contribution statement

Xianlong Zhang: Conceptualization, Writing - original draft. **Guoliang Li:** Supervision, Writing - review & editing. **Di Wu:** Software, Formal analysis. **Bin Zhang:** Writing - review & editing. **Na Hu:** Writing - review & editing. **Honglun Wang:** Writing - review & editing. **Jianghua Liu:** Writing - review & editing. **Yongning Wu:** Supervision.

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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