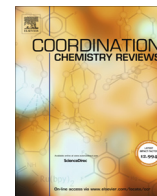




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Review

Metal–organic frameworks for biosensing and bioimaging applications

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ABSTRACT

Metal–organic frameworks (MOFs) are a class of hybrid materials assembled from metal ion/cluster connecting points and organic bridging ligands. On the basis of their properties of structural flexibility, porosity, controllable synthesis and flexibility of the pore size/wall modification, MOFs have shown great potential applications in biological sensing and imaging applications. Compared with inorganic nanomaterials (e.g., graphene, graphene oxide, gold nanoparticles and MoS_2), MOFs are potentially advantageous for biological applications owing to their intrinsic biodegradability and the ability to use biocompatible building blocks. Recently, many types of MOFs were applied for sensing DNA, RNA, enzyme activity and small-biomolecules, as well as for magnetic resonance imaging (MRI) and computed tomography (CT), which are useful techniques for clinical diagnosis. In this review, I aim to present the recent progress achieved in MOF research for biosensing and bioimaging. The compositional tenability of MOF imaging platforms should greatly facilitate their further development for clinical translation.

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Contents

1. Introduction	139
2. MOFs for in vitro biosensing/sensing	140
2.1. DNA and RNA sensing	140
2.2. Enzyme-activity sensing	143
2.3. Small-biomolecule sensing	144
2.4. Other sensing	147
3. MOFs for intracellular or in vivo imaging	147
3.1. Intracellular bioimaging	147
3.2. MR imaging (MRI)	148
3.3. Computed tomography (CT)	151
3.4. MRI and CT dual-mode imaging	151
4. Summary and future outlook	153
Acknowledgement	153
References	153

1. Introduction

Biosensing and bioimaging have been proved as promising tools to achieve rapid, sensitive, and selective analysis for clinical applications. The increasing demands for accurate sensing have signifi-

cantly promoted the design and fabrication of functional materials. A variety of organic or inorganic materials, including metallic nanoparticles, graphene oxide (GO), silica nanoparticles, quantum dots, and metal–organic frameworks (MOFs), have been employed as platforms for designing biosensors [1–4]. Among them, MOFs with specific functional sites have stimulated great interest in the area of biosensors because of their diverse structures and multifunctionalities, enabling specific molecular recognition [5–9].

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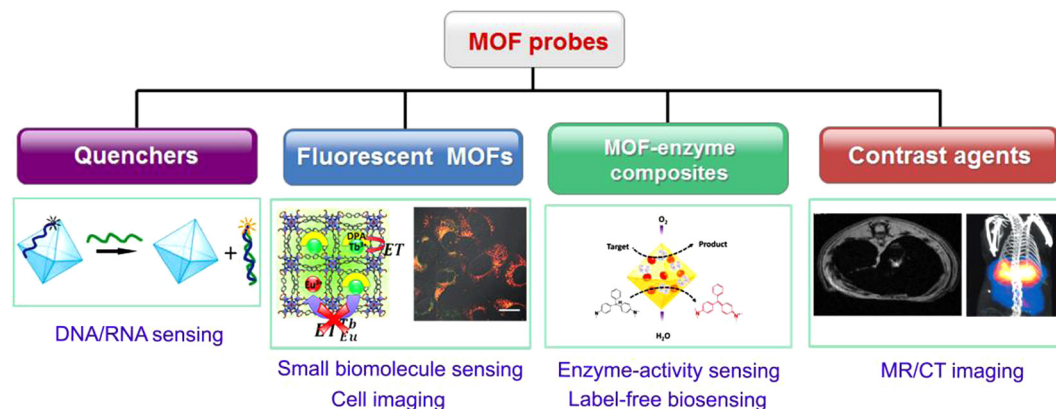


Fig. 1. Metal-organic frameworks (MOFs) for biosensing and bioimaging applications.

MOFs, also known as coordination polymers (CPs) or coordination networks, are a class of hybrid materials that consist of inorganic connectors and organic linker molecules. The acronym “MOF” was first introduced by Yaghi and co-workers in 1999 [10]. In recent two decades years, MOFs have gained considerable attention in gas storage, chemical sensing, heterogeneous catalysis and biological applications, due to their properties of structural flexibility, porosity, and controllable synthesis [7,11–16]. Particularly, MOFs are very promising for fabricating fluorescent or luminescent sensors, because the fluorescence can be generated from the metal and the ligand units, and can also be tuned by the interplay/interactions among the building components [17,18]. For example, the organic ligands containing aromatic or conjugated π moieties can give rise to fluorescent emission upon irradiation; and the metal ions (lanthanides, inorganic clusters, etc.) can also contribute to photoluminescence [19,20]. It has been proved the guest molecules can be loaded on MOFs and can induce the change of fluorescence or luminescence intensity. Therefore, a great deal of effort has been focused on designing MOF-based probes for sensing gases, explosives, solvents and biomolecules [21,22].

Many MOFs have been reported with low cytotoxicity, which is important for eventual in vivo applications [18]. If the size of MOF crystal is scaled down to nanoscale, the formed nanoscale MOFs (NMOFs) can be used as in vivo imaging agents. Furthermore, the NMOFs are intrinsically biodegradable, making it possible to rapidly degrade and clear after the intended task is completed. These specific merits of the NMOFs have enabled them as ideal platform for biosensing and bioimaging. As shown in Fig. 1, the MOF-based biosensing and bioimaging can be classified into the following ways:

- (1) MOFs are used as quenchers of fluorophores, such as using NMOFs to quench dye-labeled DNA probes for detecting DNAs or adenosine in living cells [23,24];
- (2) The fluorescence intensity of MOFs is affected by surrounding target biomolecules [25,26];
- (3) Label-free sensing of biomolecules by assembling enzymes and MOFs [27,28];
- (4) MOFs are used as contrast agents for highly effective imaging modalities, including magnetic resonance imaging (MRI) and computed tomography (CT), which are useful techniques for clinical diagnosis [13].

The advantages and challenges of MOF-based sensors have been well summarized by several previous reviews [13,18,20,21,29–31]. These extensive overviews show the wide range of MOF-based sensing applications. In recent years, the emerging functional

MOFs have shown great advantages in the field of biosensing and bioimaging. Therefore, in this review, I aim to focus on the recent research progress on the developments of MOFs for biosensing and bioimaging. Furthermore, the opportunities and some critical challenges in this field are also addressed. It is hoped that this review will inspire broader interests and will stimulate more exciting developments in biosensing and bioimaging field.

2. MOFs for in vitro biosensing/sensing

MOFs have been extensively studied as new imaging probes in the field of biomedical sensing. The MOF-based biosensing can be mainly classified as the following two approaches: (1) MOFs are used as fluorescence quenchers toward the fluorophores of analytes based on fluorescence resonance energy transfer (FRET) [32], photoinduced electron-transfer (PET) [33] or charge transfer [24]; (2) Intentional preparation of MOFs combining fluorescence or luminescence property, which can sensitively respond to their local environment or guest species [29]. The characteristic data of the MOFs for biosensing/sensing applications discussed in the present article are shown in Table 1. The MOFs for biomolecule (DNA/RNA and enzyme) sensing were mainly discussed. Additionally, MOFs for sensing some small molecules (such as glucose, dopamine, amino acids, reactive oxygen species, etc.) which play important roles in physiological process were also presented.

2.1. DNA and RNA sensing

Conventionally, the fluorescent detection of nucleic acids can be achieved on platforms, including GO, single-walled carbon nanotubes, carbon and gold nanoparticles (GNPs), as universal “quenchers” [34,35]. Recent researches have shown that MOFs also exhibit good fluorescence-quenching ability, as well as selective adsorption affinity toward single-stranded DNA (ssDNA) versus double-stranded DNA (dsDNA). In 2013, Chen and co-workers have demonstrated for the first time that the MOF $\text{Cu}(\text{H}_2\text{DTDA})$ (Table 1) is effective and reliable for the detection of HIV-1 DNA sequences and thrombin [36]. Since then, they have investigated different MOFs for sensing virus DNA or RNA. For example, they recently use the ligand $\text{H}_3\text{CmdcpBr}$, $\text{H}_3\text{CbdcpBr}$ and H_2dcbbBr to prepare a series of water-stable MOFs: $[\text{Cu}_3(\text{Cmdcp})_2(\text{dps})_4(\text{H}_2\text{O})_4(\text{SO}_4)]_n$, $[\text{Dy}(\text{Cmdcp})(\text{H}_2\text{O})_3] \cdot (\text{NO}_3) \cdot 2\text{H}_2\text{O}$, $[\text{Zn}(\text{H}_3\text{Cbdcp})_2] \cdot \text{H}_2\text{O}$ and $[\text{Cu}(\text{dcbb})_2]_n$ (Table 1) [33,37–39] Fig. 2 shows that the $[\text{Cu}_3(\text{Cmdcp})_2(\text{dps})_4(\text{H}_2\text{O})_4(\text{SO}_4)]_n$ can form electrostatic, π -stacking, and/or hydrogen-bonding interactions with two different carboxyfluorescein (FAM)-labeled probe ss-DNA. The fluorescence of

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