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A comprehensive review on luminescent metal–organic framework detectors

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ABSTRACT

Metal-organic frameworks with structures including metal ions and organic connectors have shown vast applications like catalysis, gas storage, drug delivery, and detection. In this review, applications in the detection of metal ions, explosives, pesticides, and antibiotics by luminescent metal–organic frameworks is reviewed. Metal-organic frameworks have diverse topology, pores of various sizes and shapes, vast surface area, and high number of active sites which make them more suitable for chemical detectors as compared to traditional detectors. The spectrum of this review covers antibiotics detection because their misuse has significant environmental and health consequences. This review also covers pesticide residues detection which are toxic for organisms as well as the environment. Metal contamination is another major problem, their detection is very much required and is also addressed in this review. Luminescent metal–organic framework (LMOFs) were reported to be materials for detecting various explosive chemicals. The ease of use, adaptability and tunable photoluminescence capabilities make them excellent detectors.

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1. Introduction

Metal-organic frameworks or MOFs are composed of organic ligands and metal clusters linked by coordination bonds, have gotten much attention in recent times. Pore geometry that is highly controllable and ultrahigh porosity, resulting in widespread use in molecular separation, catalysis, gas storage, drug delivery, and detection, are just a few examples. Luminescent metal-organic frameworks (LMOFs) belong to particular MOFs categories, detecting Turn off or Turn on responding [1]. Because of their customizable structures, distinctive optical characteristics, and a much longer emitted wavelength, metal–organic frameworks with luminescent properties have recently attracted much interest as excellent materials for detection systems. A considerable amount of work has been done on several types of LMOFs with good sensing properties. The majority of these frameworks used lanthanides or transition metals and have shown outstanding detection performance [2,3].

Because of the highly poisonous and non-decomposable nature of pesticides and antibiotics, these are accumulated in the environment over time, causing concern about food safety and environmental conservation [3]. LMOFs, which are fluorescence sensors, may offer a novel way to detect antibiotics with high sensitivity, quick response times, and excellent recyclability [5]. Various antibiotics like chloramphenicol (CAP), tetracycline, oxytetracycline, nitrofuran/nitroimidazole and other antibiotics are detected by LMOF's. Traditional detection methods have many drawbacks, notably more excellent prediction prices, extensive preocedure and equipment [5]. Because of their variable structure, shape, pore size, huge surface, organic frameworks of metal attracted much attention in adsorption and are promising chemical detectors for detecting pesticides [6].

Luminescence-based sensors are typically exceedingly sensitive, with the ability to detect a metal ion and low cost, ease of use, adaptability, and tunable photoluminescence capabilities. Cations and anions of metals are bridged with multi-dentate ligands with appropriate fluorescence structures in a coordinated manner to form LMOFs. The luminous qualities of LMOFs are determined by pore area, material properties, and interaction between reactant and guest the LMOFs and targeted reactant-like ions of

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metals [7]. Explosives or explosive-like hazardous chemicals are linked to concerns about human health, national security, industrial productivity, and environmental protection. Because explosives and explosive-like hazardous chemicals pose such a threat, developing sensitive, accurate, and cost-effective detection technologies is critical [8]. LMOFs were reported to be materials for detecting various explosive chemicals when it comes to explosives that are specifically targeted [9]. Fig. 1 depicts detection applications of LMOF's.

2. Detection by a luminescent metal–organic framework

2.1. Detection of antibiotics

Antibiotics are frequently utilized in daily lives and can be used to treat various diseases in the hospitals. But the antibiotic's misuse has significant environmental and health consequences, particularly in drug-resistant microorganisms. As a result, detecting antibiotics is critical in a variety of industries [11]. The 3d metal–organic frameworks having luminous properties were primarily used in sensing aqueous solutions because of their highly selective, sensitive, and lasting features. Cd-3,5-di(imidazol-1-yl) pyridine-based three-dimensional structure with stable fluorescence features was considered useful substrate for constructing fluorescent sensors for detecting antibiotics [11]. The chloramphenicol antibiotic was detected using a polymer deposited on an organic framework of zirconium with luminous properties [12]. A new luminous lanthanide metal–organic framework sensor reported detecting oxytetracycline (OTC) and tetracycline swiftly. According to a research, the presence of other species not affected the detection [13]. The Terbium ion and 1,3,5-benzenetricarboxylate based compound detected nitrofuran antibiotics as reported in a study [14]. The solvothermal approach was utilized to synthesize metal-organic framework and used for detecting nitrofuran/nitroimidazole antibiotics [15]. Recently, a research developed an easy and efficient method for detecting antibiotic residues [16]. By quenching luminescence in an aqueous solution, the luminous Zn based metal-organic framework showed good stability towards the water and was used to detect 2,4,6-trinitrophenol [17]. LMOFs were also reported to be used to detect medicines like chlortetracycline, tetracycline, ofloxacin, doxycycline, and cefixime [18]. $[Zn_2(TDA)_{2(}azopy)_2]DMF_n$ based luminous chemosensor synthesized from 4,4′-azobispyridine (azopy) and thiophene-2,5-dicarboxylic acid (H_2tda), was used to detect nitroaniline sensitively with low detection limits [19]. $Tb_2(COO)_4$ subunits were developed in an organic framework having Terbium with excellent quenching constants and less limit of detection. For dimetridazole and metronidazole gives a quick and high fluorescence response to nitroimidazole antibiotics [20]. Eu metal based MOFs detected the antibiotics furazolidone, nitrofurazone and nitrofurantoin as reported in a study [21]. A 3-Dimensional MOF, $[EuCd_{1.5}(L)_2(H_2O)_3]{\cdot}2H_2O$, was able to detect 1-Methyl-5-nitro-1H-imidazol-2-yl) methylcarbamate and 4-nitro-Niphen in the solution having water [22]. Table 1 enlists LMOF's and the antibiotics detected.

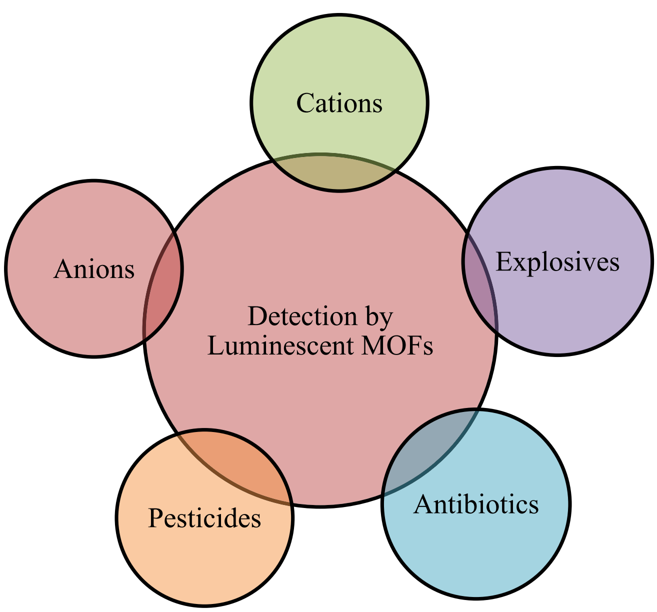

Fig. 1. Detection applications of LMOF's.

2.2. Detection of pesticides

The use of pesticides on a large scale has a significant impact on agricultural productivity. However, pesticide residues have negative health consequences. Pesticide detection has become critical regarding human health, public safety, and environmental preservation. Luminescent MOFs can detect pesticides easily due to their wide variation of pore size, emission of strong fluorescence, and large specific surface area [23]. Adsorption and determination methods aided by multifunctional MOFs have always been the highlight. MOFs are better because of their simplicity, ease of use, low cost, and ability to be employed on significant and industrial levels [24]. Water-stable LMOFs based on zirconium clusters were used to detect parathion-methyl in the aqueous phase. The Zr metal-based LMOF were employed for detecting parathion-methyl in aqueous solutions because of its strong absorption ability and bright luminescence. [25].

Organochlorine pesticides (OCPs) are commonly utilized in agriculture. Though pesticides can efficiently cure pest-induced plant diseases, their widespread use has caused significant environmental and human health harm. An organic framework having zinc metal was reported in one of the study [26]. New lanthanide MOFs with high water stability were used as multi responsive luminescence detectors sensing the pesticide dicloran [27,28]. An organic framework containing zinc metal showing luminous prop-

Table 1
LMOF's and antibiotics detected.

LMOFs	Antibiotics detected	Ref.
MIP/Zr-LMOF	Chloramphenicol (CAP)	[13]
MOF-76	Nitrofuran antibiotics	[15]
Zn-MOF $[Zn_2(IDS)(bipy)_{1.5}]$	2,4,6-Trinitrophenol	[18]
$[Zn_2(TDA)_2(azopy)_2]$	Nitroaniline	[20]
Tb-MOF	Nitroimidazole	[21]
Eu-MOF $\{[Eu(H_2O)(BTCTB)]{\cdot}2H_2O\}_n$	Nitrofurantoin (NFT), nitrofurazone (NFZ) and furazolidone (FZD)	[22]
$[EuCd_{1.5}(L)_2(H_2O)_3]{\cdot}2H_2O$	Ronidazole (RDZ) and 4-nitrophenol(4-NP)	[23]

erties was developed to detect parathion pesticide among fifteen types of organophosphates selectively. The sensor responded linearly to parathion concentrations ranging from 5 g/L to 100 g/L. The sensor's repeatability and reproducibility were 97.23 per cent and 93.76 per cent, respectively. Furthermore, throughout fifteen days, the light sensor displayed very good stability [29].

2.3. Detection of metal cations

Due to the utilization of wastewater in agriculture, including commercial and artificial wastes, heavy metal pollution of water and soil has been an issue since the rapid rise of modern industry. Both food safety and public health are in jeopardy. As a result, the metal contamination because of non-biodegradability has attracted strong focus. The thiol group builds up in proteins and enzymes in living organisms, resulting in cell dysfunction and, as a result, causing a slew of health problems in the kidneys, brain, and central nervous system. Hg^{2+} cation was detected by a framework produced by the reaction of 5-aminoisophthalic acid and nitric acid in the water, dimethylformamide, and dioxane [30]. According to a luminescence titration tests, the Fe^{3+} ions were detected by two organic frameworks of Zn and Ni metal, based on 1,4-bis(3-pyridyl)-2,3-diaza-1,3-butadiene and 1,2-phenylenediacetic acid [31,32]. A thermally stable framework of Cu metal also reported to detect Al^{3+} ions and showed luminescence properties [33]. The fluorescence experiments were used to investigate the sensing behaviour of an organic framework of zinc. When it comes to Zn^{2+} ions detection, selectivity, start concentration, detection limit, reaction time, and regeneration were all studied. A linear response to Zn^{2+} ions was established with a limit of detection of 0.7 mol/L over a concentration limit of 50–200 M. The luminescence intensification and restoration cycle was repeated on multiple occasions with no notable change insensitivity [34]. MOFs having transition metals, specifically those with the nd^{10} electronic configuration, are sensitive [35]. Cr^{3+} ions were detected using Mg-MOF of formula $[Mg(ATDC)(H_2O)_2]_n$ and were highly selective and sensitive for different metal ions. [36]. Using three-dimensional MOFs the parent metal-organic framework pores were injected with Eu^{3+} cations, resulting in Eu^{3+} entrapped products. Eu^{3+}based organic framework showed high selectivity and sensitivity towards Ag^+ ions [37]. A Terbium-based organic framework was used to develop a sensitive detection fluorescent turn-on sensor in which the Aluminium, chromium, and iron in + 3 oxidation state were present [38]. A new LMOF with nitrogen-rich nanocage was identified and used to detect $Fe^{3+}$and Cu^{2+}Ions [39]. Table 2 depicts different types of cations detected using LMOF's. A heterometallic Zn(II)–Ba(II)–organic framework having luminous property was used to detect Cu^{2+} and Fe^{3+} ions [40].

2.4. Detection of anions

Traditional methods for detecting anions are inefficient and inaccurate, and they take a long time to complete. Luminescent MOF sensors have several advantages over traditional detectors, easy to use, excellent sensitivity, remarkable selectivity, and quick reaction are just a few of the benefits. The significant characteristics of MOFs were crystal structure disintegration, ion exchange, and strong framework ions interactions [32]. Chromate anions have been widely used due to growing urbanization and industrialization. Human diseases are easily accumulated in organisms, so it is important to have chemical detectors for sensing $CrO_4^{2-}/Cr_2O_7^{2-}$ which are being developed to be more accessible, efficient, and cost-effective. Organic frameworks of cadmium ($[Cd_3(BTC)_2(TPT)(water)_2]{\cdot}4H_2O$ (H_3BTC = trimesinic acid, TPT = tris(4-pyridyl)triazine) were used to detect CrO_4^{2-} and $Cr_2O_7^{2-}$ions. Importantly, following the sensing investigations, these MOF can be easily and quickly produced, suggesting great recyclability for sensing CrO_4^{2-} and $Cr_2O_7^{2-}$ ions. As a result, these were employed as a regenerable multi-responsive luminescence sensors to detect harmful compounds [41]. The 3D luminous europium (III) MOF was prepared successfully and was acknowledged as a highly rapid with sensitivity dual luminous chemosensor to detect PO_4^{3-} ions [42]. Luminous metal–organic frameworks (LMOFs) development for specific analytes is challenging due to not having understanding about the analytes' sensory processes and methodologies for appropriate framework-matching [43].

2.5. Detection of explosives

Explosives detection is critical to ensure the safety of the public. Detection is a usual method, but it necessitates the use of large instruments. The microporous MOFs with luminous characteristics have emerged as explosive detection materials. They are also desirable because of their size, functionality, ease of use, and structure adjusted accordingly during synthesis. Nitroaromatic explosives, like 2–4-6 trinitrotoluene (TNT), are dangerous to health and the environment, so detection is necessary. The fluorescent 8-hydroxyquinoline zinc was added to the imidazole framework-8 of zeolite and this framework showed change in colour from ivory to light red when exposed to TNT [45]. Isoreticular metal–organic framework (IRMOF-3) particles with an amine group were also employed as a TNT sensing material. IRMOF-3 particles were made with the metal precursor zinc nitrate and the 2-aminoterephthalic acid. 3-carboxyglutaric acid and $C_2H_4(NH_2)_2$ were the C and N precursors for N-doped carbon quantum dots respectively. In N-doped carbon quantum dots, the intensity of emission of photoluminescence of IRMOF-3 particles at 430 nm was considerably increased [46]. Cd-based MOF showed excellent compatibility, selectivity, and reproducibility as a multipurpose luminous sensor and was stable even after ten repetitions of detecting nitro explosives [47]. In the aqueous phase, a Zirconium based MOF showed good explosive detection capabilities [48]. $[Zn(L)]{\cdot}2MeOH{\cdot}H_2O$ was the first metal–organic framework based on 8-hydroxyquinoline. By coupling oxyquinoline ligand H_2L with 2-COOH groups with Zn, $2MeOH(H_2O)$ was created to detect nitroaromatic explosives [51].

3. Conclusion

Luminescent metal-organic frameworks can detect wide range of analytes due to their structural diversity, large tunability, and unique optical property. Several research papers targeted detection by LMOF's which are summarized in this review. The effect of components or structures on LMOF sensing properties is studied extensively for detecting metal cations and anions, explosives, pesticides, and antibiotics. LMOFs were used to detect medicines like chlortetracycline, tetracycline, ofloxacin, doxycycline, and cefixime. These can efficiently detect various antibiotics like chloramphenicol, tetracycline, oxytetracycline, nitrofuran and nitroim-

Table 2
LMOF's and metal cations detected.

LMOFs	Cations detected	Ref.
ZnAPA	Hg^{2+}	[32]
Cu(I)-MOF $[CuI(BPDPE)]_n$	Al^{3+}	[35]
$Zn_2(BDC)_2(DABCO)$	Zn^{2+}	[36]
Mg-MOF $[Mg(ATDC)(H_2O)_2]_n$	Cr^{3+}	[38]
[LCU-103]	Fe^{3+}, Cu^{3+}	[41]
Eu^{3+}@UIO-67	Ag^+	[39]
Tb–TCPP	Al^{3+}, Cr^{3+}, and Fe^{3+}	[40]

idazole antibiotics. Metal ions are also detected with selectivity by luminescent metal-organic frameworks. Specially MOF ([Cd_3(-BTC)$_2$(TPT)(water)$_2$]·4H_2O detect efficiently and selectively CrO_4^{2-}, $Cr_2O_7^{2-}$, and PO_4^{3-} ions. IRMOF-3 framework are also capable of sensing nitroaromatic explosives, like 2–4-6 trinitrotoluene (TNT). Zirconium based MOFs are also very efficient in detection explosives. The detection efficiency of LMOFs as chemical sensors is mainly driven by synergistic interactions among distinct subsets, such as guest molecules, inorganic, organic component properties, and so forth. The combination of luminescence sensing and other functionalities will broaden LMOFs applications in various areas, notably medical diagnosis and therapy and photoelectric devices. So LMOF's proved to be very efficient and selective in detection of various substances due to its vast properties. With partnerships amongst different subjects, functionalized LMOFs will have a brighter future.

CRediT authorship contribution statement

Gunjan Gupta: Conceptualization, Visualization, Investigation, Methodology, Writing – original draft. **Archana Thakur:** Data curation.

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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