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REVIEW

A Review on Metal-Organic Frameworks: Synthesis and Applications

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Metal-organic frameworks (MOFs) are inorganic-organic hybrid porous materials that are composed of positively charged metal ions and organic linkers. The metal ions form nodes that connect the arms of the linkers together to form one-, two-, or three-dimensional structures. Due to this void structure, MOFs have an unusually large internal surface area. They have received enormous interest in recent years particularly as newly developed porous materials. They possess a wide range of potential applications like gas storage, catalysis, sensors, drug delivery, adsorption, *etc*. In present review article, synthetic methods and applications of MOFs have been discussed.

Keywords: Metal-organic frameworks, Synthesis techniques, Gas storage, Sensors, Catalysts, Drug delivery.

INTRODUCTION

Metal-organic frameworks (MOFs) were first discovered in the year of 1965. MOFs are a class of compound in which metal centers or clusters and multidentate organic groups as linkers [1-4] are coupled together by coordinate bonds to form one-, two- and three-dimensional highly porous structures [5] as shown in Fig. 1. So, metal-organic frameworks (MOFs) are inorganic-organic hybrid porous materials. In general, MOFs are called as coordination polymers. However, for 2D and 3D networks the term MOF is more appropriate than coordination polymer. Thus these are also called as porous coordination polymers (PCPs) [6,7]. Several coordination geometries such as octahedral, tetrahedral, trigonal-planar, *etc*. and also interesting structural architectures are obtained by varying the coordination number of metal ions. Transition metals, lanthanides, actinides, *p*-block elements, alkaline earth metals and even mixed metals are usually employed for the formation of MOFs. The N-containing aromatics or multivalent aromatic carboxylic acids are commonly used as organic linkers in MOFs.

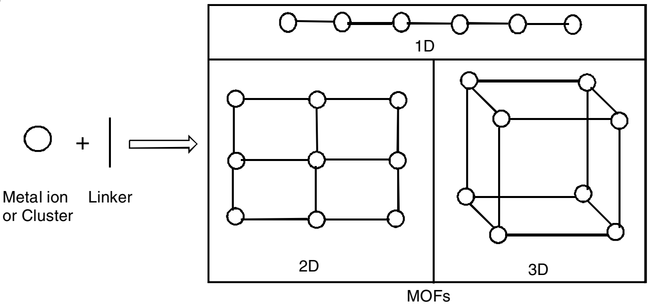

Fig. 1. Schematic representation of a MOF structure

Over the last two decades, a lot of research work has been done on MOFs. Due to structural flexibility, high surface area, small density and tunable pore size, MOFs have a wide range of potential applications in the field of gas adsorption and storage, separation, catalysis, sensing, molecular recognition, drug delivery, non-linear optics, luminescence, *etc*. [8-18]. Present article provides a review of literature on synthesis and application of MOFs.

Connectors and linkers: For the formation of metal-organic frameworks metal ions (connectors) and organic compounds (linkers) have been used. These are called "Primary Building Units" (PBUs). In some MOFs, metal-oxygen-carbon clusters are applied instead of metal ions alone. These metal-oxygen-carbon clusters are termed as "Secondary Building Units" (SBUs). Generally, 1st row transition metal ions like Cr^{3+}, Mn^{2+}, Fe^{3+}, Co^{2+}, Ni^{2+}, Cu^{2+}, Zn^{2+} are used as connectors in the formation of MOFs [19-35]. In addition, various alkali metal ions [36,37], alkaline-earth metal ions [38-40] and rare earth metal ions [41-46] have been applied as a connector for the synthesis of MOFs. For the production of MOFs in various synthetic process, chloride, sulphate, nitrate, perchlorate, acetate, oxide of metals have frequently used as precursor. Nevertheless, metal rods are used for the electrochemical synthesis of MOFs.

Organic compounds which are used as linkers also called as bridging ligands. They efficiently bridges two or more metal ions by their donor atoms. Generally, N-donor (amines, nitriles), O-donor (carboxylates, phosphates), S-donor (sulphonates) ligands have been used for the synthesis of MOFs. Linkers behave as electron pair donor whereas metal ions act as electron pair acceptor. Thus organic ligands can easily donate its lone pair of electrons to the vacant orbital of the targeted metal ions to form coordination bonds and ultimately MOFs with proper geometry are produced. The structures of different linkers used for the production of MOF materials are shown in Fig. 2.

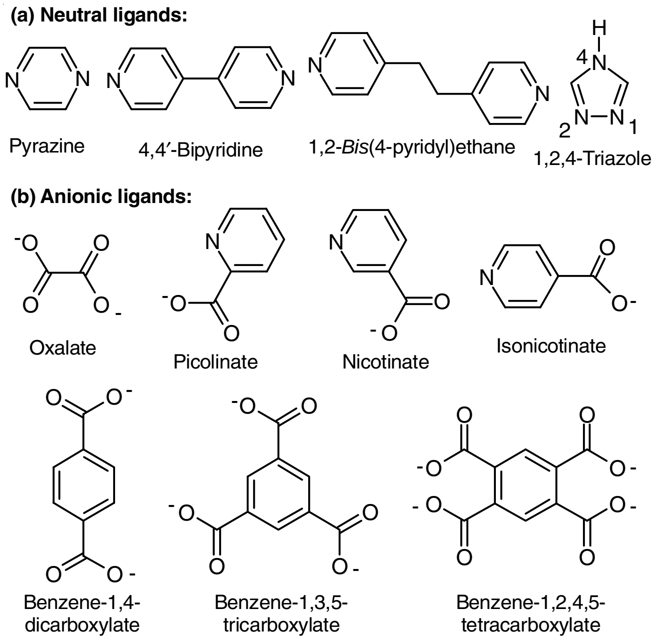

Fig. 2. Some ligands used in synthesis of MOFs

Synthesis of metal organic frameworks: Numerous procedures have been employed for the synthesis of MOFs. These can be happen in liquid phase, where at first solutions of metal salt and ligand are prepared separately using solvent and then mixed together or solid salt and ligand are mixed then solvent is poured to the mixture to get a solution. Now as the solvent plays a crucial role for the development of MOFs having various features like redox potential, reactivity, stability constant, solubility, *etc*. so the choice of solvent is most vital. However, products depend upon the variety of parameters, such as temperature, reaction time, pressure, pH and solvent, must be considered as well.

$$\text{Metal ions + Organic Linkers} \longrightarrow \text{MOFs}$$

To produce MOFs different synthetic methods like slow evaporation, hydro (solvo) thermal, microwave, electrochemical, mechanochemical and sonochemical are presented here.

Slow evaporation method: It is a conventional method for the preparation of MOFs, where no external energy is required. In this process, the starting materials (metal salts and ligands) are dissolved by solvent or by mixture of solvents. Then the solution was kept at room temperature in an inert atmosphere, the solvent will be slowly evaporated and solution gets concentrated. After a few days, single crystals will appear with proper shape and size. The key factor for the preparation of MOFs is the solubility of the compounds. To increase the solubility, mixture of solvent may be used. As the whole process was carried out at room temperature, so this is a lengthy process. Murinzi *et al.* [47] successfully synthesized a new 1D metal organic framework constructed by cobalt(II) as metal salt and 2,6-pyridinedicarboxylic acid as linker using this method. The cobalt(II)-MOF acts as a catalyst and its electrocatalytic properties were studied. Dzhardimalieva *et al.* [48] prepared a new MOF, $Cu_3(BTC)_2.2DMF\cdot 2H_2O$ at room temperature. The Cu-MOF is used as a sorbent to remove organic dyes.

Hydro (solvo) thermal method: The most conventional and universally used method for the preparation of MOFs is hydro (solvo) thermal method as it affords different morphologies. In this method, a solution is prepared taking the metal salt and organic linker in a solvent or a mixture of solvents. Commonly used organic solvents are dimethyl formamide (DMF), diethyl formamide (DEF), acetonitrile, acetone, ethanol, methanol, *etc*. Subsequently the solution obtained is introduced in glass vials (for lower temperature) or in Teflon-lined autoclaves or bomb reactor (for temperature higher than ~ 400 K) [48]. When, H_2O was used as solvent then the process is called hydrothermal. A number of coordination complexes have been successfully synthesized with the help of this method.

Zhou *et al.* [49] synthesized a new Ni-MOF using $Ni(NO_3)_2\cdot 6H_2O$ and 4,4′-biphenyldicarboxylic acid (BPDC) under hydrothermal route. Ni-BPDC is applied as a electrode material. Ranjbar *et al.* [50] reported a manganese metal organic framework constructed by $Mn(NO_3)_2$ as metal salt and three ligands (i) 2,6-pyridine dicarboxylic acid, (ii) 1,2,3-benzene tricarboxylic acid and (iii) 1,3,5-benzene tricarboxylic acid *via* this method. The CO_2 and SO_2 gas uptake capacity was also investigated using Brunauer-Emmett-Teller (BET) analysis. Guo *et al.* [51] synthesized a new Co-MOF, $[Co_2(4\text{-}ptz)_2(bpp)(N_3)_2]_n$ {where, ptz = 5-(4-pyridyl)tetrazole, bpp = 1,3-bi(4-pyridyl)propane and NaN_3} The Co-MOF shows important catalytic activity for the oxidation of hydrazine and reduction of nitrobenzene. Wu *et al.* [52] prepared two new metal organic framework *viz*. (i) Zr-MOF and (ii) Pd-MOF by hydrothermal technique and investigated their photocatalytic activity. Qin *et al.* [53] successfully synthesized two novel MOFs $\{[Zn_2(L1)(tp)(formate)_2]\cdot H_2O\}_n$ (where (L1 = 1,2-*bis*{2,6-*bis*-[(1*H*-imidazol-1-yl)methyl]-4-methylphenoxy}ethane, H_2tp = terephthalic acid) and $\{[Cd_2(L2)(ip)_2]\cdot 2H_2O\}_n$ (where L2 = 1,3-*bis*{2,6-*bis*[(1*H*-imidazol-1-yl)methyl]-4-methylphenoxy} propane), H_2ip = isophthalic acid) by hydrothermal method. They show potential luminescent properties and particularly Zn-compound has adsorption property for C_{60} molecule.

Microwave method: In this technique, microwave irradiation is the source of energy for reaction. Here, the reaction is occurred on the basis of interaction between electromagnetic waves and mobile electric charges, such as polar solvent molecules or ions in the solution. This method was generally used in organic chemistry during last few decades but nowadays it is applied for the preparation of MOFs. The quality of the product is similar as that of the normal solvothermal process. This is a high speed synthetic route [54,55]. Besides, this method has various advantages like high efficiency, phase selectivity, particle size reduction and morphology control [56-61]. Lin *et al.* [62] synthesized two nanoscale metal-organic frameworks (NMOFs) *viz.* (i) $[Mn(BDC)(H_2O)]_2$ and (ii) $[Mn_3(BTC)_2(H_2O)]_6$ {where BDC = terephthalic acid and BTC = trimesic acid} successfully by microwave-assisted syntheses. Bae *et al.* [63] synthesized a zinc MOF *i.e.* $[Zn_2(NDC)_2(DPNI)]$ {where NDC = 2,6-naphthalenedicarboxylate, DPNI = N,N′-di-(4-pyridyl)-1,4,5,8-naphthalene tetracarboxydiimide]} using microwave method.

Electrochemical method: In this method, the target metal ions are relentlessly supplied through anodic dissolution as a metal source in place of metals salts into the reaction mixture where it reacts with the dissolved linkers and electrolytes [64] and formed the desired MOFs. HKUST-1 is first MOF, which was synthesized by researchers at BASF in year 2005 using electrochemical method [65]. This synthetic procedure has been extensively used for the large scale production of MOFs in industries. The main advantages of this process are avoiding use of metal salts, lower reaction temperature and particularly rapid synthesis, than conventional solvothermal synthesis. For example, Zn and Al containing MOFs, ZIF-8, MIL-100(Al), MIL-53(Al) and NH_2-MIL-53(Al) have been synthesized by Gascon *et al*. [66] adopting the electrochemical method. They also reported that the yield of the product may vary with the temperature, solvent, electrolyte and current-voltage density. Li *et al.* [67] successfully prepared the fluorescent MOF of $[Zn_3(btc)_2]$ with 1,3,5-benzenetricarboxylic acid as linker and Zn electrode and used to identify the nitro explosives.

Mechanochemical method: In mechanochemical method, the chemical reaction is carried out with mechanical force. The most important advantage of this process is that no need of organic solvent to carry out the reaction. Generally, metal oxides are used as starting materials instead of metal salts in this method. Mechanochemical synthesis is simple, economical and environment friendly. Alammar *et al.* [68] have been synthesized three luminescent metal organic frameworks (MOFs), $[Ln_{0.5}Gd_{0.5}\{C_6H_3(COO)_3\}]$ with a MIL-78 structure, where Ln = Eu, Tb and Dy by this method without using organic solvent simply and rapidly taking metal carbonates and benzene 1,3,5-tricar-boxylic acid, commonly called as trimesic acid. Recently, modified mechanochemical synthesis *i.e.* liquid-assisted grinding (LAG) has been fruitfully applied for the fast production of MOFs by Beldon *et al.* [69] where the reaction mixture is prepared by adding a minimum amount of solvent.

Sonochemical method: In this synthetic process, ultrasonic radiation (20 kHz-10 MHz) is applied to the solution of reaction mixture and as a result some chemical or physical changes are observed into the molecules and ultimately MOFs having novel morphologies and unique properties are achieved. In the reaction medium high temperatures and pressures are produced by the ultrasonic radiation. So within a short period of time, reaction is completed and we get crystals with proper size and shape. This method is very fast, reproducible, economical and environment friendly [70]. Masoomi *et al.* [71] prepared two Zn(II) based MOFs by sonochemical method and the compounds are characterized by PXRD, SEM and IR spectroscopy. Morsali *et al*. [72] synthesized 3D Zn based MOF, $[Zn(OBA)(DPT)_{0.5}]\cdot DMF$ sonochemically taking 4,4′-oxy*bis*-benzoic acid (H_2OBA) as linker and 3,6-di(pyridin-4-yl)-1,2,4,5-tetrazine (DPT) as spacer. Lestari *et al.* [73] synthesized one Zn based MOF $[Zn_3(btc)_2]$ by sonochemical as well as electrochemical method and compared between the two process. They reported that higher yield and smaller size particles were generated by sonochemical method than the electrochemical method.

Applications: MOFs have a wide range of applications like gas storage, catalysis, sensor, drug delivery, *etc*. Some major applications are illustrated as follows:

Gas storage: The main application of MOFs is gas storage due to their exceptional large surface areas, tunable pore structure, low weight, open metal centers and functionalized polar groups. The numbers of MOFs, which have been examined for H_2 storage are about 300. Ma & Zhou [74] studied porous MOFs as H_2 and CH_4 storage and CO_2 capture. Wong-Foy *et al.* [75] reported MOF-177, which is produced by taking 4,4′,4″-benzene-1,3,5-triyltribenzoate (BTB) and $[Zn_4O]$ clusters, best example of H_2 uptake material. It exhibits a gravimetric H_2 uptake of 7.5 wt% at 70 bar and 77 K, due to its high BET surface area (~5000 $m^2\ g^{-1}$) and large pore volume (1.59 $cm^3\ g^{-1}$). MOF-5 (IRMOF-1) [76], a Zn-based MOF which have 3800 $m^2\ g^{-1}$ BET surface area, capable of takes up 7.1 wt% of H_2 at 40 bar and 77 K. In addition, several well known H_2 storage MOFs are MIL-101, MOF-210, MOF-205, HKUST-1, PCN-12, NU-100 and NOTT-102 [77-79]. The large surface area of MOFs is the main driving force for greater interaction amid metal ions and hydrogen molecules. For this reason, a class of compound can easily uptake the H_2 molecules.

Sensing: Photoluminescence property was shown by maximum number of MOFs because each aromatic part of the linker gets excited by absorbing UV-visible light and give luminescence. MOFs having luminescent properties [80] are generally used for the development of different types of sensors. Qian *et al.* [81] reported various MOFs, which act as potential sensor materials. Several luminescent MOFs have been used for temperature sensing material. For example, Zn-based MOF $[Zn_3(TDPAT)(H_2O)_3]$ (TDPAT = 2,4,6-*tris*(3,5-dicarboxylphenylamino)-1,3,5-triazine) gives ligand centered emission band at 435 nm upon excitation at 370 nm. The emission band remains unchanged with decreasing temperature but the intensity increases due to cooling restrictions of the thermally activated intramolecular rotations and non-radioactive decay [82]. Tb-dmbdc (dmbdc = 2,5-dimethoxy-1,4-benzene dicarboxylate) compound also exhibits temperature dependent luminescence property [83].

Catalysis: The catalytic activity of these materials is observed owing to the existence of unsaturated metal centres or catalytic species existing in the pores or intrinsic sites in the MOFs. The knowledge of catalysis is very essential for the improvement of sustainable chemical processes [84] in chemical industry. MOFs have been significantly utilized as heterogeneous catalyst because of their tunable porosity, high surface area and diversity in metal ions and organic linkers. The MOFs act as a good catalyst when the metal ions are not wholly covered by organic ligands or when the coordinated water molecules are eliminated giving a vacant coordination position on the metal centre. For instance, in $[Cu_3(btc)_2]$ (btc = 1,3,5-benzene tricarboxylate) MOF, after removing the coordinated aqua ligand vacancy is created on Cu atom upon thermal activation [85]. Many organic reactions have been catalyzed applying nanoporous MOFs. Such as, Knoevenagel condensation reaction is catalyzed by means of either $[Cd(4\text{-btapa})_2(NO_3)_2]$ [86] (btapa = 1,3,5-benzene tricarboxylic acid tris[N-(4-pyridyl)amide]) or $[Cr_3F(H_2O)_2O(bdc)_3]$ [87] (bdc = 1,4-benzenedicarboxylate). Zou *et al.* [88] synthesized a three dimensional MOF taking an isolated metal-organic cubic building block $[Ni_8L_{12}]^{20-}$ (H_3L = 4,5-imidazoledicarboxylic acid) bridged by alkali-metal ions (Na^+). This MOF showed stable catalytic activity for the oxidation of CO to CO_2. Few examples of MOFs catalyzed reactions are listed in Table-1.

Drug delivery: MOFs have been used as drug delivery materials since their properties can be adapted by changing the coordinated groups of the frameworks and by delicately tuning the pore size [102]. The family of MIL [103] composed of trivalent metal ions and different bridging carboxylates are suitable for drug delivery due to their greater pore sizes (24-25 Å) and related surface areas (3100-5900 $m^2 g^{-1}$). For example, MIL-100 and MIL-101 have been examined for delivery of drug (ibuprofen) as they have discrete structure and controlled porosity. Serre *et al.* [104] established encapsulating drug molecules (ibuprofen) in MOFs (chromium carboxylate), MIL-100 and MIL-101, which exhibit drug storage capacities of 35 and 140 wt%, respectively and drug release activities of 5 to 6 days under physiological conditions. MOFs of iron(III) carboxylate *e.g.*, MIL-8, MIL-88A, MIL-100 and MIL-101 can easily capture antitumor, antiretroviral and anticancer [105] drugs. A new MOF composite $Fe_3O_4/Cu_3(BTC)_2$, produced by taking Fe_3O_4 nanorods and $Cu_3(BTC)_2$ (HKUST-1) nanocrystals, adsorbed an anticancer drug namely Nimesulide of about 0.2 g/g of composite and it completely released after 11 days [106].

Electrochemical applications: In addition to the various applications like drug delivery, gas storage and catalysis, MOFs exhibit another important application, which is utilized in our daily life. MOFs have been used as electrode materials for the production of lithium ion battery to store energy. Lithium ion batteries are produced by using an anode electrode, electrolyte and a cathode electrode. As it is known that the lithium ion batteries are widely used in numerous electronic devices like cell phones, robots, computers, laptop, electric cars, *etc.* due to their fantastic properties such as light weight, high energy density and low pollution [107,108]. Recently, lithium sulfur batteries [109] are developed and also utilized in electrical equipment and aerospace systems.

The first reported MOF, $[Zn_4O(BTB)_2](DEF)_m(H_2O)_n$ (BTB = 1,3,5-benzenetribenzoate, DEF = diethylformamide) (MOF-177) has been applied [110] as an anode material for storage of Li. This MOF is irreversibly decomposed to form metallic zinc by the following reaction:

$$[Zn_4O(BTB)_2](DEF)_m(H_2O)_n + e^- + Li^+ \longrightarrow Zn + Li_2O$$

Another Zn-based MOF, $[Zn_3(HCOO)_6]$ (HCOO = formate) [111] can reversibly changed to lithium formate by lithiation. This reaction is the driving force to improve the long-term cyclability.

$$Zn_3(HCOO)_6 + 6Li^+ + 6e^- \rightleftharpoons 3Zn + 6HCOOLi$$

$$3Zn + 3Li^+ + 3e^- \rightleftharpoons 3LiZn$$

This MOF has a steady capacity of around 560 mAh g^{-1} at 60 mA g^{-1} when the voltage range is 0.005-3.0 V, up to 60 cycles. Tarascon *et al.* [112] reported that $[Fe^{III}(OH)_{0.8}F_{0.2}\text{-}(O_2CC_6H_4CO_2)]$ ($O_2CC_6H_4CO_2$ = 1,4-benzenedicarboxylate) (MIL-53(Fe)) can be used as a cathode material. Its reversible capacity is 70 mA h g^{-1} within 1.5-3.5 V. The applications of different MOFs are listed in Table-2.

Conclusion

Recently, metal-organic frameworks (MOFs) have become more and more applicable in chemical industries. Although

TABLE-1
FEW EXAMPLES OF MOFs CATALYZED REACTIONS

MOF	Reaction(s) catalyzed	Ref.
$[Cd(bpy)_2(NO_3)_2)]$	Cyanosilylation of aldehyde	[89]
[Co(BPB)]	Oxidation of olefin	[90]
$[Cu(2\text{-pymo})_2]$ and $[Co(PhIM)_2]$	Aerobic oxidation of olefin	[91]
$[Co(sal)(H_2O)(Py)_3]$	Epoxidation of olefin	[92]
$[Cu(bpy)(H_2O)_2(BF_4)_2(bpy)]$	Ring-opening of epoxide	[93]
[In(OH)(hippb)]	Acetalization of aldehyde	[94]
$[Cd_3Cl_6L1_3]$	Alkylation of aldehyde	[95]
ZIF-8	Cycloaddition of CO and epoxides	[96]
$[Zn_4O(bdc)_3]$ and $[Zn_4O(ndc)_3]$	Friedel Crafts alkylation	[97]
MIL-100(Fe)	Friedel Crafts benzylation	[98]
$[Pd(2\text{-pymo})_2]$	Oxidation of alcohol	[91]
MIL-101(Cr)	Heck coupling/Knoevenagel condensation	[99]
$[Ag_3(tpha)_2]BF_4$	1,3-Dipolar cycloaddition	[100]
UiO-66	Cyclization of citronella	[101]

TABLE-2
SELECTED APPLICATIONS OF METAL-ORGANIC FRAMEWORK

MOF	Synthetic route	Application	Ref.
Zr-CAU-24	Hydrothermal	Detection of aflatoxin B1	[113]
UMCM-151	Hydrothermal	Adsorption of light hydrocarbons	[114]
Mn(II)-MOF	Solvothermal	Adsorption of dyes	[115]
$[Zn_2(NDC)_2(DABCO)]_n$	Hydrothermal	CH_4/CO_2 gas separation	[116]
ZIF-8	Ultrasonic assisted	Removal of tetracycline and oxytetracycline antibiotics	[117]
ZIF-12	Hydrothermal	Rn adsorption	[118]
Ru_3-NHC-MOF	Solvothermal	Catalytic hydrogenation of CO_2 to formic acid	[119]
$[In_6O_3Tb_3O(CBDA)_3].18DMF\cdot 3H_2O$	Solvothermal	Gas adsorption	[120]
$MIL\text{-}101\text{-}NH_2$	Hydrothermal	Catalyst for the synthesis of 3-aryl-2-oxazolidinones	[121]
Co-MB	Hydrothermal	Photocatalysis for hydrogen production	[122]
ZnNi MOF@ZnO	Hydrothermal	Photoelectrochemical water oxidation	[123]
Ni(Fe)-MOF	Solvothermal	Electrocatalytic water oxidation	[124]
CUP-1-Ni	Hydrothermal	Electrocatalytic water oxidation	[125]
MOF-808	Hydrothermal	Biocatalyst	[126]
Fe_2Co_1 MOF-74	Hydrothermal	Removal of arsenic	[127]
MIL-53(Al)-GO	Hydrothermal	Removal of arsenic	[128]
UiO-68 NMOF	Hydrothermal	Biocatalytic decomposition of the duplex capping units	[129]
Cu/Tb@Zn-MOF	Hydrothermal	Sensor for detection of aspartic acid	[130]
$[Co(NPDC)(bpee)].DMF\cdot 2H_2O$	Solvothermal	Luminescent sensing for MnO_4^- and Hg^{2+}	[131]
$Zr\text{-}UiO\text{-}66\text{-}N_2H_3$	Solvothermal	Fluorescent sensor for phosphate and 4-nitrobenzaldehyde	[132]
UiO-66	Solvothermal	Electrolyte additive for Li-metal battery	[133]
Zn-POMCF	Electrochemical	Lithium-ion batteries	[134]
Li-UiO-66(MOD)	Mechanochemical	Lithium sulfur battery	[135]
$Cu_2(CuTCPP)$	Solvothermal	Lithium-sulfur battery	[136]
MOF5DC	Hydrothermal	Sodium-ion battery	[137]
Ni-MOF	Hydrothermal	Supercapacitor	[138]
CoCuNi-bdc	Hydrothermal	Supercapacitor	[139]
Co-MOF	Hydrothermal	Supercapacitor	[140]
$[Fe(C_6N_6O_2)(H_2O)_4]$	Solvothermal	Proton-exchange membrane fuel cells	[141]
CD-MOF	Vapour diffusion	Drug delivery	[142]
NU-1000 and NU-901	Hydrothermal	Drug delivery	[143]
$[Dy(HABA)(ABA)](DMA)_4]$	Solvothermal	Drug delivery	[144]
CD-MOF-1 and CD-MOF-2	Solvothermal	Drug delivery	[145]
$[Gd(BCB)(DMF)](H_2O)_2$	Solvothermal	Drug delivery	[146]

various synthetic methods and a huge number of MOFs with diverse arrangements of metal ions and linkers are reported, their current applications have great attention to the researchers. MOFs exhibit enormous applications *e.g.*, catalysis, gas storage and separation, adsorption, magnetism, sensors, electrical energy storage, drug delivery systems, *etc*. Some of these are very helpful to our everyday life. In this review article, various synthetic methods of MOFs along with their applications have been discussed. Several new applications will appear as the research theme, which becomes progressively admired.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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