

REVIEW PAPER

Review on Synthesis Method, Classification and Adsorption Separation and Sensing Application of Metal-Organic Frameworks (MOFs) Based Composites

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ABSTRACT

A group of supramolecular solid materials known as “metal-organic frameworks” (MOFs) is a type of hybrid network made up of a variety of inorganic and organic linkers that are all tightly bonded to metal ions. These classes of compounds have a larger surface area with the benefit of variable pore sizes, a diverse structure, and a lovely appearance. They are promising materials for a range of applications because they are easy to develop and have consistent, fine-tunable pore structures. The controlled mixing of MOFs with functional materials is resulting in the development of new multifunctional composites and hybrids that display unique properties that outperform those of their parts as a whole. The structural characteristics, classification, and The most widely used and successful strategies for MOF composite synthesis are presented in the encapsulation method, Solvothermal method, Solution impregnation, and Click chemistry (reaction) method), numerous characterization techniques and their applications MOFs composite have all been covered in this review. Crystals with extremely high porosity and good thermal and chemical stability can be produced by carefully choosing the MOF ingredients. Because of these properties, MOF composites can be used for a wide range of applications, including, sensing toxic chemicals, drugs, gases, trace metals, components of foods, and many more, and also for the detection of different materials. This is a rapidly developing interdisciplinary and novel research area therefore to present the current situation of the field; this article has covered recent achievements as well as new avenues to investigate the future scope and uses of MOF composites/hybrid.

Keywords: Metal-Organic Frameworks; MOF composites; Porous material; Separation; Sensing

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INTRODUCTION

Metal-organic frameworks, often known as MOFs, have become a diverse family of crystalline materials with extremely high porosity (up to 90% free volume) and massive inner surface areas, exceeding 6 000 m²/g [1]. Since its initial definition in the 1990s, Metal-Organic Frameworks (MOFs), which consist of coupling units (metal ions

or metal-oxo clusters) coordinated by organic ligands, have drawn a lot of interest [2, 3, 4]. Due to its potential for separation [5], adsorption [6], catalysis [7], chemical sensors [8], miniature electronics [9], optical materials [10], drug delivery [11], electrical and optoelectronic devices [12], and other applications, MOFs have gained significant attention by researchers. The three most commonly used orders of 3D porous covalent polymers

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are zeolites [13], metal-organic frameworks (MOFs) [14], and covalent organic frameworks (COFs) [15, 16]. Metal-organic frameworks are crystalline porous materials having regular network topologies made of metal ions (or metal clusters) and organic linkers. They are also known as porous coordination polymers (PCPs) [17]. MOFs may be produced utilizing a variety of unique synthetic techniques, including slow diffusion, hydrothermal (solvo-thermal) [18], electrochemical [19], mechanochemical [20], microwave-assisted heating [21], and ultrasound [22], depending on the final structures and features. The composite of MOF can be made in a variety of ways, including, encapsulation [23], impregnation [24], infiltration [25], solid grinding [26], coprecipitation [27], click reaction [28], and more. Diverse preparation techniques can result in different MOF composite characteristics, which can increase the usefulness of composite materials [29, 30]. Numerous MOFs have been produced, and they have many beneficial properties, including (a) adjustable pore size; (b) ordered structure; (c) tunable diameter; (d) resistance to changes in their morphologies; (e) convenient and affordable processing conditions; (f) easy sample collection; (g) resistance to metal aggregation; (h) resistance to architecture disintegration; and (i) attachment of other substances inside the pores or on the surface [31, 32, 33]. Even though MOFs have a lot of benefits, they also have significant drawbacks that restrict their use, including low mechanical strength, poor chemical and hydrothermal stability, and a single function [34, 35]. As a result, MOF composites were designed to address these shortcomings. In comparison to pure MOFs, MOF composites display novel chemical and physical properties or improved performance because of the synergistic interaction between MOFs and functional materials [36, 37]. When compared to pure MOFs and other conventional materials, the use of MOF composites provides significant advantages and development potential examples according to Olorunyomi et al. report MOF composites are identified as advanced materials with potential for deployment in analytical devices for chemical and biochemical sensing applications [38], Ahmed and Jhung also reported the MOF composites adsorption applications [39] and Li et al. state that because of MOF-based membranes' greater performance, they have garnered a lot of interest in separation applications [40].

CLASSIFICATION OF MOF AND MOF COMPOSITE

There are various kinds of MOFs, and these MOFs can integrate with other functional resources to develop MOF composites with a variety of functions (Fig. 1). Examples of these materials include metal NPs [41], metal oxides [42], quantum dots, carbon materials [43], molecules, polyoxometalates [44], polymers [45], and enzymes [46]. Depending on their constituent parts, the MOFs can be divided into a variety of categories. According to the functional materials that have been doped, the MOF composites are categorized.

Classification of MOFs

Iso-reticular MOFs (IRMOFs)

Reticular chemistry, a fundamental technique used to produce these kinds of materials, involves the connecting of molecular-scale building blocks into predetermined shapes that are recognized and ordered by strong bonding forces [47]. The primary building blocks of IRMOFs are secondary inorganic structural units $[Zn_4O]^{6+}$ and various aromatic carboxylic acid ligands [48]. Isorecticular MOFs (IRMOF), a subclass of MOFs, have a similar cubic topology to MOF-5 and are connected by linear organic linkages [49]. Due to its easy synthesis, effective gas adsorption, and useful storage properties, IRMOF-n ($n = 1-16$) has been the subject of significant research [50, 51].

Zeolitic imidazolate frameworks (ZIFs)

Zeolitic imidazolate frameworks (ZIFs) are a distinct family of metal-organic frameworks (MOFs) that maintain the basic chemical connection over wide variations in the network topology and related attributes. Because of their huge potential for hydrogen storage and carbon dioxide capture, ZIFs currently draw a great deal of attention [52]. There are different produced ZIFs including ZIF-5, ZIF-7, ZIF-8 [51, 52], ZIF-9 [53], ZIF-11, ZIF-71, ZIF-67 [54], ZIF-90 [55], ZIF-L [56], ZIF-95 [57], ZIF-100 [58], etc. Tetrahedral $Si(Al)O_4$ units are chemically bonded by bridging O atoms to form more than 150 various forms of framework that makeup zeolite structures. In water, aqueous alkaline solution, and refluxing organic solvents, ZIFs exhibit exceptional chemical stability [59].

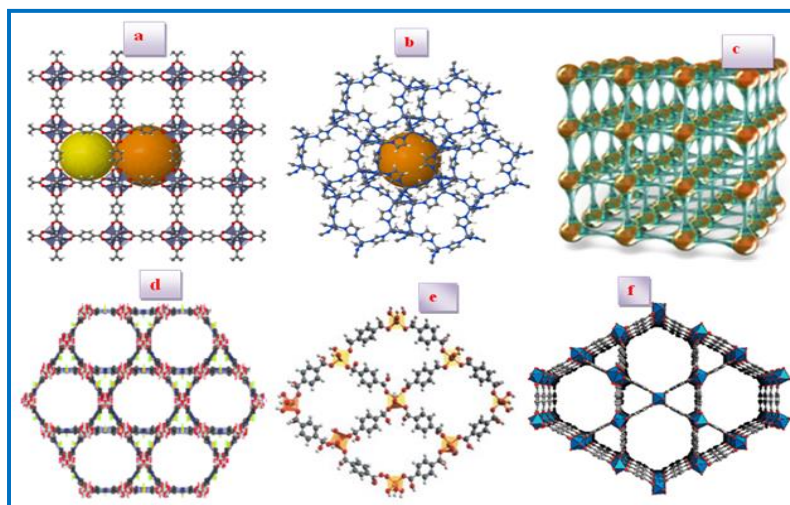


Fig. 1. Structure of metal framework of a) IRMOF-5 [49] b) ZIF-8 [52] c) PCPs [61] d) PCN-222 [66] e) MIL-88B (Fe) [75] f) MIL-68 (In) [84]

Coordination polymers (PCPs)

PCPs also known as Metal-organic frameworks (MOFs), are porous, polymeric (1D, 2D, or 3-D) materials linked together by coordination bonds between metallic ions and organic linkers [60]. In comparison to zeolite or activated carbon, PCPs have a better application potential as a result of their uniformity, permeability, large surface area, and performance factor. In addition to coordination bonds, PCPs also contain other weak contacts or electrostatic interaction bonds (H-bonds, π -electron stacking, or van der Waals interaction), which gives them the potential to change their structural composition which is advantageous in separation applications. The use of Zn_4O -based MOF compounds for fuel gas storage is widely recognized due to their high porosity and huge surface area [61].

Porous coordination networks (PCNs)

These are a novel class of nanoporous materials that possess good chemical and mechanical durability, huge surface areas, high porosities, and variable pore sizes [62]. Some examples of PCN-10, PCN-11 [62], PCN-12, PCN-14, PCN-15 [63], PCN-333 [64], PCN-224 [65], and PCN-222 [66], materials widely used as a sensor and for H_2 storage due to their nanoscopic cages. It has been determined that several PCNs, including PCN-5, PCN-6 [67], PCN-610, PCN-46, PCN-68, PCN123, and PCN-124, are promising for CO_2 removal [68, 69, 70]. Results show that PCN-59 exhibits the highest CO_2/N_2 selectivity

among the various PCN frameworks taken into consideration [71].

Materials Institute Lavoisier (MILs)

MILs are porous metals of carboxylate salts with large pores and permanent porosity made up of various trivalent metal cations and carboxylic acid ligands [72]. MILs can serve as vehicles for drug delivery. Busulfan, azidothymidine triphosphate, doxorubicin, and cidofovir are examples of anti-tumor and anti-AIDS active compounds that could be collected by MIL series frameworks and returned in human organs such as the liver [73]. MIL materials include MIL-53(Fe) [74], MIL-88B (Fe) [75], MIL-68(Ga) [76], MIL-88A (H_2O_2) [77], MIL-125(Ti) [78], MIL-101(Cr) [79], Fe-MIL, Al-MIL [80], Cr-MIL [81], etc. Due to its permeable or flexible structure, MIL-53(Fe) is frequently used to remove pigments and hazardous chemicals from aqueous solutions [82]. An electrochemical sensor that can accurately detect the presence of PQ in a food sample was built using Fe_2O_3 -MIL-100 [83].

Classification of MOF composites

MOF-metal nanoparticle composites

Due to their critical role in catalysis, metal nanoparticles (MNPs), especially small-size MNPs, are increasingly gaining significant attention. Unfortunately, Small MNPs are thermodynamically unstable and more likely to agglomerate during catalytic reactions because of their high surface energy, which results in a loss of efficiency [85]. MOFs are applied to modify these

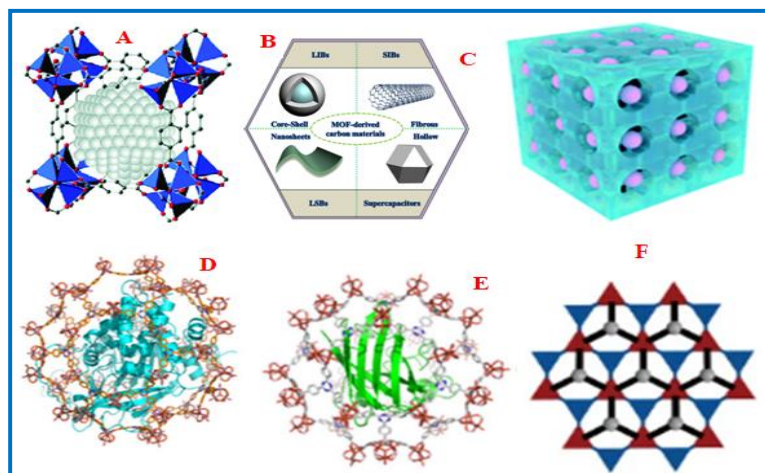


Fig. 2. The structure and MOF- derived materials and their composite a) Ru@MOF-5 composite [89] b)MOF-derived carbon energy materials [116] c) metal oxide@ KIT-6 [117] d) Enzyme encapsulation on FNPCN-333-SOD e) Enzyme encapsulation on FNPCN-333-CAT [118] f) MOF/COF composite [119].

conditions. There are two alternative methods for producing MNPs inside MOFs. The primary and most popular technique involves using MOFs as stabilizing substrate components, which provide restricted areas that prevent agglomeration and control particle size. The process requires precursor infiltration in stages, followed by decomposition or reduction [86, 87]. The other technique involves encapsulating organic material, surfactants, and polymers that have stable prefabricated NPs in solution, followed by the addition of appropriate precursors to produce the framework around the prepared NPs [88]. The MNPs- MOF compounds formed using those methods including Pd NPs/MOFs [89], Au NPs/MOFs [90], Ru NPs/MOFs [91], Cu NPs/MOFs, Pt NPs/MOFs [92], Ni NPs/MOFs [93], Ag NPs/MOFs [94], etc are some examples (Fig. 2). High-performance anodes for lithium-ion batteries can be made using Si-MOFs [95]. Cu-MOF-GN/GCE shows effective anti-interference characteristics for the detection of H_2O_2 and AA in the presence of different carbohydrates [96].

MOF-carbon composites

Their large surface area, electrical properties, and high stability of carbon-based materials (CMs) make them excellent candidates for use as catalysts, energy storage systems, waste disposal, and drug delivery systems [97]. Because of their drawback in the field of sensing, separation, and adsorption MOF-carbon composite materials have received attention in recent times. As an example, smaller molecules like ammonia are challenging to remove

from airstreams using activated carbon. Many findings confirmed that MOF-carbon composites are interesting materials for use in air purification [98]. The Co-MOF-MPC composite may be very promising for the designing of electrochemical environmental sensors because it shows excellent catalytic performance for the oxidation of hydrazine and reduction of nitrobenzene [99]. CeCu-MOF, synthesized by a hydrothermal process, is utilized as an electrode material in lithium-ion batteries and supercapacitors [100]. By using an in-situ growth technique, Ni-MOF@CNT material was produced on the GN substrate and used as a unique self-supporting composite for all-solid-state supercapacitors with a high energy density [101].

MOF-metallic compound composites

Metal oxide nanoparticles serve a crucial function, in the field of materials chemistry, medicine, agriculture, information technology, biomedical, optical, electronics, catalysis, environment, energy, and sensing [102]. A new technique (MOF-metallic compound composite) was developed to examine the uses and limitations of metal oxide nanoparticles. To produce composites with the best attributes, metallic compounds such as SiO_2 [103], Fe_3O_4 [104], ZnO [105], CuO [106], CdS [107], and ZnS [108] have been doped into MOF materials. The in-situ solvothermal approach was used to produce Zr-based UiO-66- NH_2 MOF for improved activity towards Cr (VI) adsorption and photocatalytic H_2 evolution [109]. Metal-organic frameworks derived

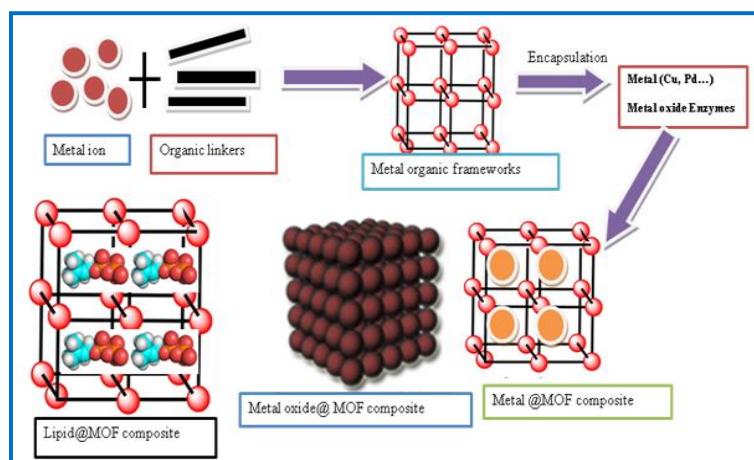


Fig. 3. Synthetic route of Metal@MOF, Metal oxide@MOF, Lipid@MOF composite synthesized by using encapsulation method.

$\text{In}_2\text{O}_3/\text{ZnO}$ synthesized using a straightforward co-precipitation process and thermal annealing in air can also be used as a highly sensitive H_2S gas sensor [110]. The solvothermal approach is applied to synthesize bimetallic FeO/NiO MOF and rGO-based composites, which can also be used in the methanol oxidation reaction [111]. The adsorbent $\text{MgFe}_2\text{O}_4@\text{MOF}$ was synthesized using a simple solvothermal approach to eliminate the organic dyes Rhodamine B (RB) and Rhodamine 6G (Rh6G) from water samples [112].

MOF-enzyme composites

Enzymes are flexible, environmentally friendly biological catalysts that have been widely used in the chemical, food, cosmetics, and pharmaceutical industries. Due to Low stability at higher temperatures and extreme pH values, slow recovery, and the challenge of reusing enzymes have all restricted its use [113]. The development of immobilization as a new technique for the stabilization, convenience of restoration, and persistent usage of enzymes. According to their method of synthesis, MOF-enzyme composites can be divided into four major categories: surface attachment, covalent coupling, pore entrapment, and co-precipitation [114]. The encapsulation of lipid into ZIF-67 MOF forms a lipase@ZIF-67 composite and this composite is used as a catalyst for the production of biodiesel. Lipid encapsulation in ZIF-67 greatly improved the enzyme's heat stability, storage stability, and reusability [115]. GOx-ZIF-8 biocomposites are an excellent material for electrochemical biosensing applications for

the detection of glucose and the composite was synthesized using a biomineralization-assisted in-situ encapsulation approach [116]. In general, the enzyme-MOF composites have greater application potential and have superior catalytic activity than free enzymes.

SYNTHESIS OF MOF COMPOSITE

Most of the problems related to everyday activities are solved by MOF, a fascinating and emerging science that is also making significant contributions to the advancement of technology. As technology develops, new synthetic techniques for MOF composites are continuously being developed. Some of the synthetic methods including mechanochemical, electrochemical, microwave, and sono-chemical methods, encapsulation, solution impregnation, solvothermal, one-pot, and diffusion techniques.

Encapsulation method

The most popular and established method for producing MOF composites is encapsulation as shown in Fig. 3. The “build-bottle-around-ship” or “encapsulation method” is based on two key processes. (I) NPs are synthesized that are homogeneous in size, structure, and morphology. (II) Building MOF shells on previously produced NPs [120, 121]. Encapsulation allows for the incorporation of different morphologies and sizes of NPs, carbon compounds, molecular catalysts, enzymes, and other substances within MOF shells [122]. Pd@ZIF-8 is a MOF-composite material synthesized by the encapsulation method in a

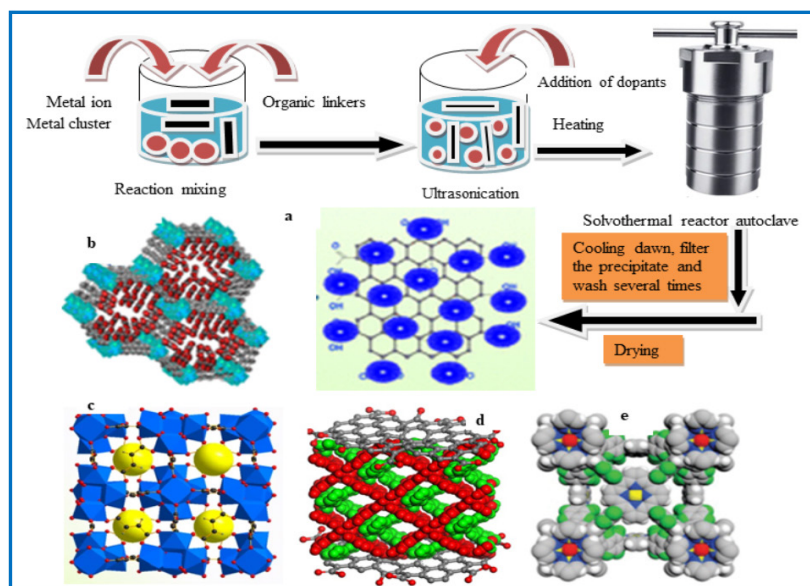


Fig. 4. Schematic diagram for the synthetic route MOF-composite materials by solvothermal methods and structure of some MOF-composites a) Cu-MOF-GO [129] b) Mg/Cu-MOF-74 [130] c) MOF-801 (Zr) [131] d) GO-TMU-16-NH₂ [132] e) PCN-624 [133].

simple, rapid, and sustainable manner [123]. EY@Zr-MOF composite was also synthesized by encapsulation of EY molecule and Pd on the surface of the ZIF-8 MOF [124].

Solvothermal method

A reaction mechanism in an enclosed system in the presence of a solvent at a temperature greater than that of such a solvent's boiling point. This technique demonstrates the advantages of low cost and effective diffusion [125, 126]. An easy solvothermal technique was used to synthesize Fe-MOF/RGO composites with various RGO ratios. The precursor used for synthesis Fe-MOF/RGO was RGO, N, N-dimethylformamide (DMF), 1, 4-dicarboxybenzene, iron (III) chloride hexahydrate, and acetic acid glacial at a temperature of 150 °C for 2 h in Teflon-lined stainless autoclave (Fig. 4) [127]. The 6% weight of the CdS/MIL-101 catalyst was synthesized using the solvothermal method. In a Teflon-lined autoclave, MIL-101, DMSO, and C₁₀H₁₄CdO₄ were combined and stirred for two hours [128].

Solution impregnation

The most popular and simple way to produce assisted catalysts for a range of heterogeneous reactions is the impregnation method [134, 135]. There are three fundamental steps. A substantial surface area support is first impregnated with a

metal precursor solution, which is then condensed at a high temperature. Finally, the metal precursor solution is reduced in a suitable environment to make the catalysts [37, 136]. An Ag@MOF-5@chitosan composite was synthesized using the solution impregnation method (Fig. 5).

chitosan and silver into MOF-5 using a solution of 1 M sodium chloroacetate with 5% sodium hydroxide, DMF, and silver nitrate solution [137]. Pd@MOF-5 was also synthesized by solution impregnation techniques. This composite was prepared by impregnating Pd into MOF-5 using palladium acetate or palladium acetylacetonate in absolute chloroform [138]. The solution impregnation is mostly used for preparing metal@MOF composite using the salt solution of the metal. The metal-MOF composite is prepared as follows in Fig. 6.

Click chemistry (reaction) methods

Sharpless and his friends first introduced click chemistry in 2001, which grew out of a desire to use molecular assembly's capabilities for as many different types of applications as feasible (Fig. 7) [140,141]. Click chemistry is defined as a class of reactions that are quick, easy to perform, simple to refine, flexible, regiospecific, and produce large amounts of output [142,143,144]. Using click chemistry synthesis method Cu based MOF-composite was synthesized (Fig. 8) [145]. Zr-based

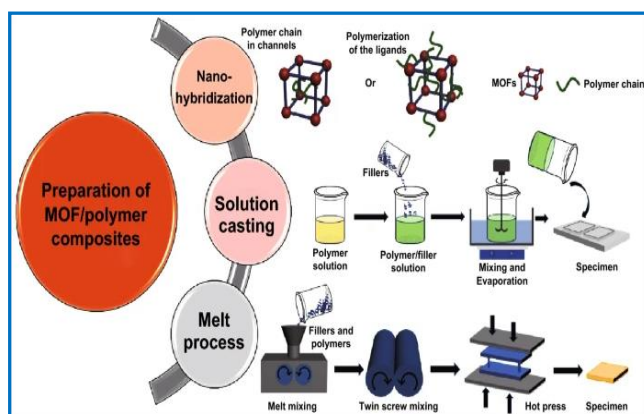


Fig. 5. Schematic of the possible methods for preparing polymer@MOF composites [139].

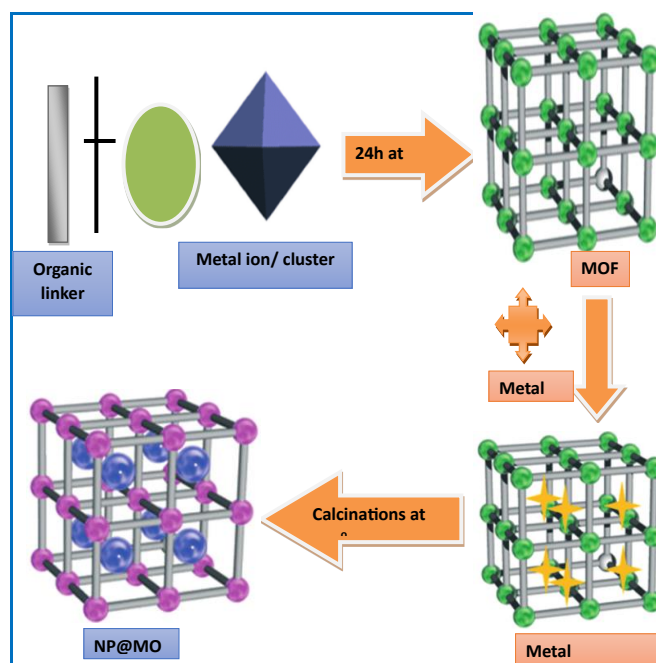


Fig. 6. Schematic illustration of synthetic strategies of nanoparticle-MOF composite [139].

MOF (UiO-68-triazole-A) was also synthesized by using a click reaction. The azide functional group in UiO-66-N₃ was modified by phenylacetylene using the standard azide-alkyne click reaction. Triazole units produced as a result serve as effective fluorescence assays for metal ions [146, 147].

Characterization of MOF composite materials

The maximum amount of energy that the material absorbed (λ_{max}), electronic transition, band gap, functional group, morphology, topology, size and shape, stability, and the surface charge of the material was analyzed using different

characterization techniques. The most commonly used instrument for characterizing MOF-composite was ICP, Solution NMR, UV-visible, FT-IR, XRD, SEM, TEM, Zeta-potential, TGA, EDS, N₂ adsorption, Cyclic voltammetry, etc. if further analysis is needed some other characterizing technique like XPS, XANES, EXAFS, SAXS spectroscopies can be applied.

UV-visible analysis of MOF-composites

From the Fig. 9 (a) below, the maximum wave length that PW11V@MIL-101 absorbed with the concentration of the composite 5 mg/L was 680

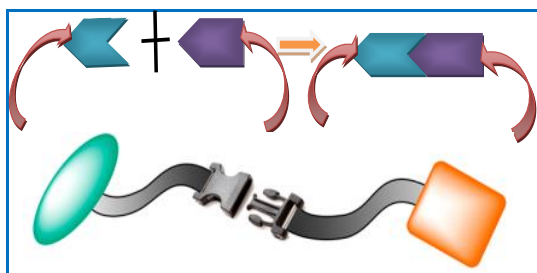


Fig. 7. Schematic diagram representing the principle of click reaction [140]

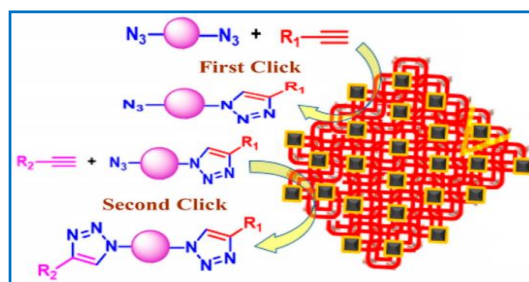


Fig. 8. Synthetic route of Cu@MOF by click chemistry for catalytic application [145]

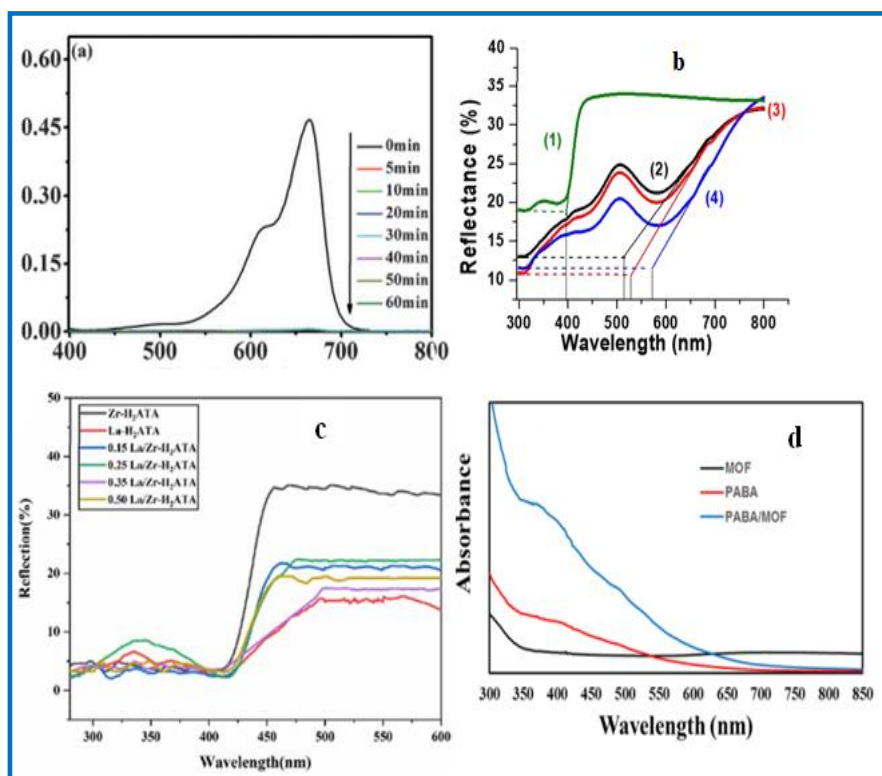


Fig. 9. UV-visible spectroscopy for a) PW11V@MIL-101[148], b) 1% TiO_2 /polymer composite, (2) 0.5%, (3) 1%, (4) 2% MIL-53(Cr)/polymer composites [149], c) UV-visible spectra of Zr- H_2BDC , La- H_2BDC , La/Zr- H_2BDC , La/Zr- H_2BDC , La/Zr- H_2BDC , and La/Zr- H_2BDC [150], d) UV-visible spectra of PABA [151].

nm [148]. (b) Shows the maximum wave length absorbed for the composite (1) 1% TiO_2 /polymer composite, (2) 0.5%, (3) 1%, (4) 2% MIL-53(Cr)/polymer composites was 396, 515, 520, 572 nm respectively [149]. (c) The maximum absorption wavelengths of Zr- H_2BDC , La- H_2BDC , La/Zr- H_2BDC , La/Zr- H_2BDC , La/Zr- H_2BDC , and La/Zr- H_2BDC were 355, 334, 361, 443, 373, and 365 nm, respectively. La/Zr- H_2BDC has a higher maximum wavelength relative to the other in the UV-visible region [150]. (d) the UV-visible spectra of PABA have a broad absorption band between

300 and 600 nm which can be attributed to the $\pi-\pi^*$ transition of the benzenoid rings and quinoid rings, respectively, for the polymer and composite [151].

SEM analysis of MOF-composites

The morphology of some of the selected MOF-composite materials is shown in Fig. 10, (a). The crystals of the original $[\text{Cu}_3(\text{BTC})_2(\text{H}_2\text{O})_3]_n$ sample are octahedral with a smooth surface and have an average size of 10 mm. (b) The surface of the magnetic MOF tends to be rougher after immobilization by Fe_3O_4 -Py [152]. (c) MIL-125(Ti)

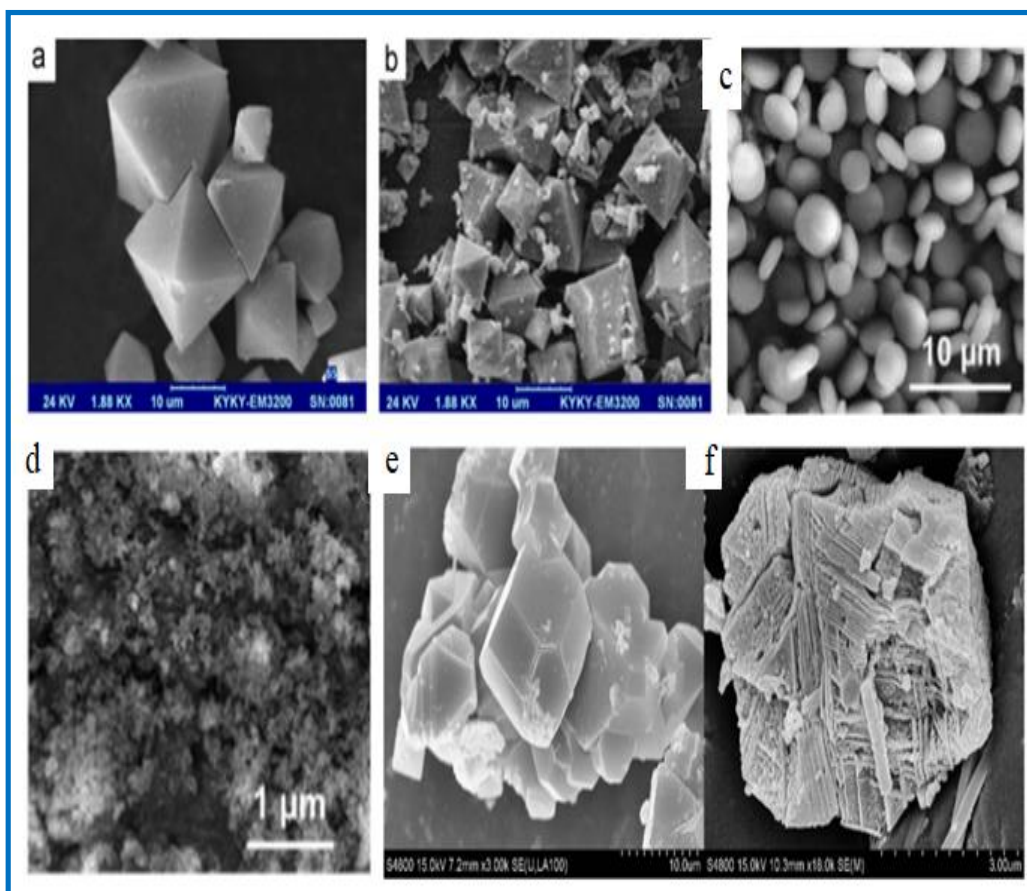


Fig. 10. SEM image for MOF-composite of a) $[\text{Cu}_3(\text{BTC})_2(\text{H}_2\text{O})_3]_n$ [152] (b) magnetic MOF [152], (c) MIL-125(Ti) [153], d) MIL-53(Al) [153], (e) $\text{Cu}_3(\text{BTC})_2 \cdot \text{H}_2\text{O}$ [154], (f) $\text{Fe}_3\text{O}_4/\text{Cu}_3(\text{BTC})_2\text{-H}_2\text{Dz}$ [154].

composite with morphology in between ovoidal or flat cubes with round corners (d) MIL-53(Al) has flat cube morphology with very small particle size [153]. (e) and (f) $\text{Cu}_3(\text{BTC})_2 \cdot \text{H}_2\text{O}$, $\text{Fe}_3\text{O}_4/\text{Cu}_3(\text{BTC})_2\text{-H}_2\text{Dz}$ has crystal losing structure [154].

XRD analysis of MOF-composites

The XRD pattern of different MOF-composites shown in Fig. 11 was analyzed as,

a) due to a highly ordered structure of Ce-MOF@TEOS material a sharp peak was shown at 28.2° . Three other diffraction peaks at 32.3° , 47.8° , and 56.6° were displayed, respectively. The pattern of Ce-MOF@TEOS and Ce-MOF are the same but the intensity of the peak in MOF@TEOS was strong indicating that the crystallite structure changed due to the presence of TEOS [155].

b) There are no identifiable CsPbBr_3 peaks in the XRD pattern of the $\text{CsPbBr}_3/\text{MOF-5}$ composite. Because of the weak crystalline structure of CsPbBr_3 PeQDs than MOF-5 and the small amount

of PeQDs in the composite, the XRD pattern of CsPbBr_3 PeQDs was covered by MOF-5. Due to this there, the XRD pattern of $\text{CsPbBr}_3/\text{MOF-5}$ moves slightly to the small angle direction [156].

c) In this the XRD peak of In_2O_3 , MoS_2 , and $\text{In}_2\text{O}_3/\text{MoS}_2$ composite was clearly seen. The characteristic peaks of In_2O_3 located at 2θ of 21.42° , 30.63° , 35.46° , 41.79° , 45.76° , 51.05° and 60.64° , which assigned to the (211), (222), (400), (332), (431), (440) and (622) planes of the cubic structured In_2O_3 , respectively. The peak for MoS_2 occurred at 2θ of 14.31° , 32.98° , 39.59° , and 58.56° which were assigned to the s (002), (100), (103), and (110) planes respectively. The XRD pattern of the composite $\text{In}_2\text{O}_3/\text{MoS}_2$ was all the peaks in In_2O_3 and MoS_2 exist. There is no additional peak shown indicating the purity of the material [157].

d) In this the peak of all the composite compounds (a, b, c) and the MOF (black) was the same. The intensity of MOF HKUST1 was strong compared to the composite this is due to the doped material

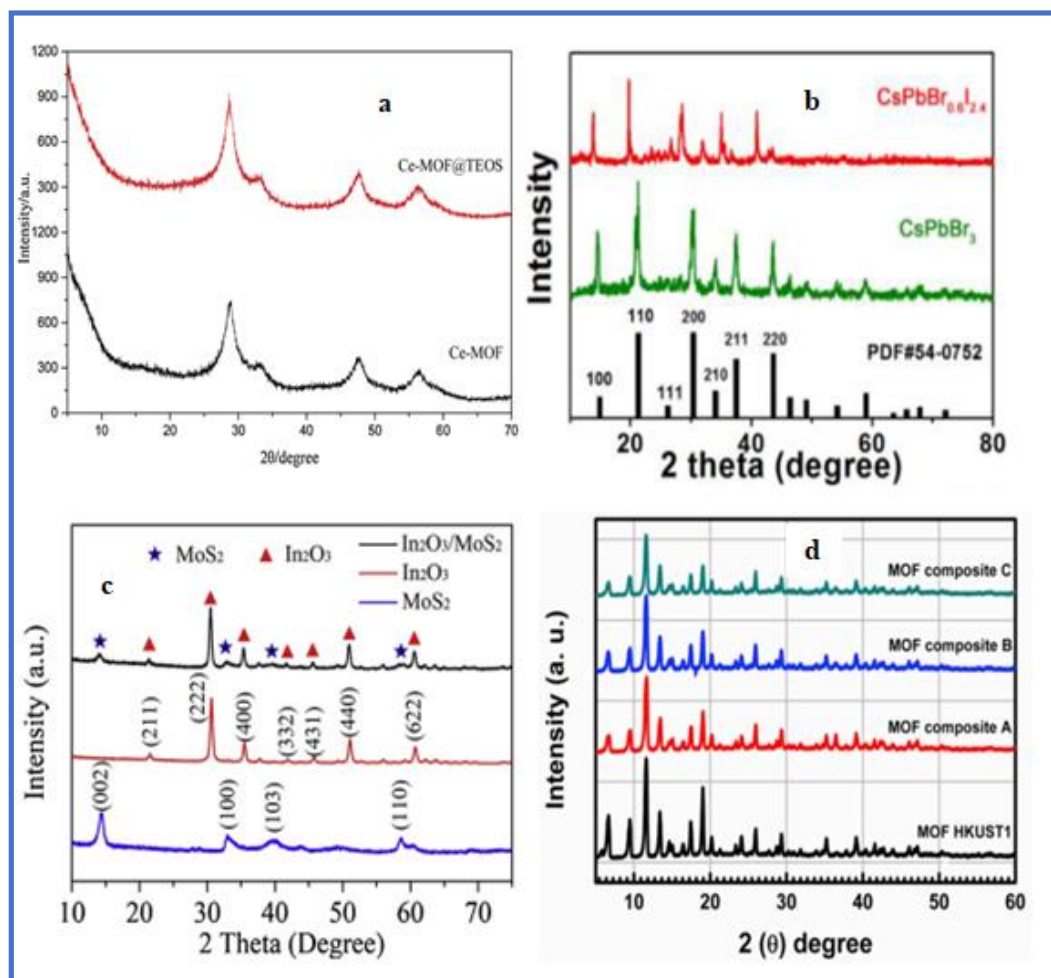


Fig. 11. The XRD pattern of a) Ce-MOF and Ce-MOF@TEOS[155] b) CsPbBr₃ PeQDs, CsPbBr_{0.6}I_{2.4} PeQDs[156] c) In₂O₃, MoS₂ and In₂O₃/MoS₂ [157]d) MOF HKUST1 (black), MOF HKUST1@ APTES NWs@ Al₂O₃(compound A, B, C) with different amount of dopants (0.2g: 1.85g, 0.4g: 2.4g, 0.6g: 4.9g of APTES NWs and Al₂O₃) respectively[158].

(APTES NW s) in the composite decreasing the crystallinity of the material [158].

Thermogravimetric analysis (TGA) of MOF-composite

From the TGA curve shown in Fig. 12:

a) the two DTG peaks below zero indicate the reaction that occurred was exothermic. The composite is thermally stable until the temperature reaches 300 °C. When the temperature reaches 300 °C the composite starts to lose its weight until the temperature becomes 440 °C due to the removal of water molecules. After that, the composite is thermally stable indicating pure Cu-MOF [159].
 b) This composite loses its mass between the temperature range between 20-100 °C and 430-580 °C due to the removal of water and volatile matter. The thermal stability indicates the purity of the

composite [160].

c) Almost the material is thermally stable in all temperature ranges indicating the material is very pure [161].

d) All the composites show weight loss but in comparison, Zr-MOF-PVA-M was less pure due to more contamination during material doping [162].

Application of MOF-composite

Because of the properties of MOF-composite like crystalline porosity, reusability, selectivity, performance, and stability, they are highly applicable for absorption separation [163]. They are mainly used for gas separation [164], oil-water separation [165], separation of linear and branched hydrocarbon [166], adsorption separation of heavy metals in water, dyes [167], and so on. MOF-

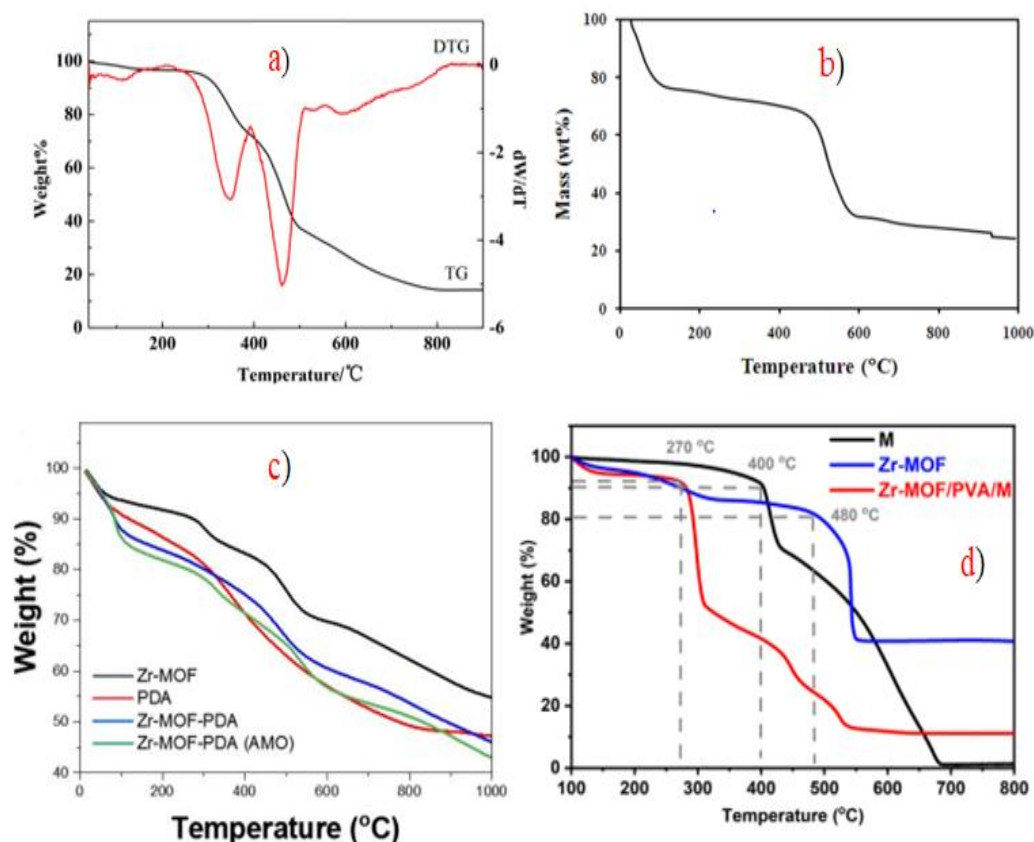


Fig. 12. TGA curve for a) Cu-MOF b) pd@MOF c) Zr-MOF, PDA, Zr-MOF-PDA, Zr-MOF-PDA (MOF) d) M, Zr-MOF, Zr-MOF-PVA/M (M is melamine)

composite has a wide application in different fields some of which are shown in Fig. 13.

Gas separation application

MOF-composite can be used for gas separation including separating CO_2 , purifying C_2H_4 , purifying C_3H_8 , separating noble gases, and separating isotopes. Mainly CO_2 is separated from N_2 , CH_4 , and C_2H_2 . Flue gas formed during combustion can cause the greenhouse effect. So highly selective MOF-composites like IL/MOF/POLYMER [168], cellulose@UiO-68- NH_2 [169], ZIF-8/BMIM/SCN [170], etc. can be used. CO_2 can be separated from methane. Methane is the most widely used chemical fuel and is the hydrocarbon with the lowest carbon content. After combustion, it contains the gas CO , CO_2 , NO , and SO_2 . Because of this MOF-composite can be used for separating those gases from methane [171]. For the manufacture of 1,4-butanediol, polymers, and polyurethanes as well as for welding and metal cutting, acetylene (C_2H_2) is a necessary precursor. During the

production of acetylene main gases including CO_2 exist as a byproduct. Removing CO_2 is challenging because of its kinetic diameter. So MOF-composite is the best method to separate C_2H_2 and CO_2 [172]. Those materials are also used to purify C_2H_4 by adsorption separation of contaminants from it. The primary steps in the purification of C_2H_4 are the separation of $\text{C}_2\text{H}_2/\text{C}_2\text{H}_4$, $\text{C}_2\text{H}_6/\text{C}_2\text{H}_4$, and $\text{C}_2\text{H}_2/\text{C}_2\text{H}_6/\text{CO}_2/\text{C}_2\text{H}_4$ [173]. Removing those impurities is the primary step in using C_2H_4 as a raw material in different industries, especially for the fabrication of polyethylene. Separating $\text{C}_2\text{H}_6/\text{C}_2\text{H}_4$ is challenging because some physical are similar like their boiling point and kinetic diameter. At this time, MOF is the best method to purify C_2H_4 [174]. The other gases purified and separated by MOF composite including $\text{C}_3\text{H}_4/\text{C}_3\text{H}_6$ [175], $\text{C}_3\text{H}_4/\text{C}_3\text{H}_4/\text{C}_3\text{H}_6$, and $\text{C}_3\text{H}_6/\text{C}_3\text{H}_8$ [176]. Purified forms of krypton (Kr) and xenon (Xe) have found extensive use in a variety of industries, including medical imaging, industrial lighting [177], insulation, lasers [178], illumination, and spaceship propellant [179]. Those

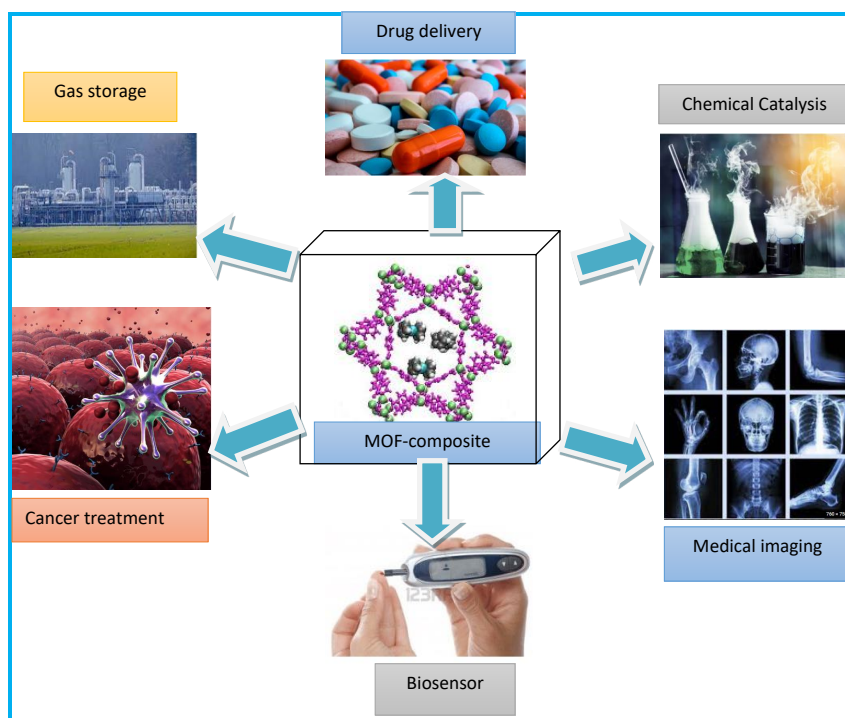


Fig. 13. Various applications of MOFcomposite

noble gases are dangerous for the environment if they are released into the atmosphere. MOF-composite is applicable for Xe/Kr separation [180]. The materials are also important to separate gases with different separations called isotopic separation. The property and usage of those gases depends on their isotope. For example, isotopes of hydrogen have their property and application. MOF is important to separate isotopic gases like D_2/H_2 mixtures [181]. Separation of linear/branched alkane hydrocarbons ($C_5-C_6-C_7$), cyclic C_6 isomers (benzene/cyclohexane), cyclic C_8 isomers (styrene/ethyl benzene, and xylene isomers), Xylene isomers, and Styrene/ethyl benzene separation was done by MOF-composite. There is a persistent risk of leakages due to the transportation and storage of oil and petroleum products as a major source of energy around the world [182]. Because of this, the water surface is polluted by that oil. To remove the oil from the water it is costly. Due to this MOF-composite is the best method to remove oil from the water surface and it is also environmentally friendly during separation [183].

Wastewater treatment application

Water is the most vital source of all life activities but the growing number of contaminants that

can pose intensified effects on both humans and aquatic life makes water pollution a more serious environmental issue [184,185]. Nowadays dyes are used in a variety of industrial sectors, such as tanning leather, paper production, textiles, and printing. For instance, azo dyes are widely used in the textile industry to generate large volumes of colored organic and inorganic wastewater. Consequently, their treatment is essential [186,187]. In 2021, Shokri reported that the synthetic polymer Polyvinyl alcohol (PVA), which is used as a binder, shaping agent, and raw material in industries such as paper, textile, pharmaceutical, and membrane industries, may cause pollution issues if improperly handled upon release into the aqueous environment [188]. Waste water typically contains not only dyes it also contains heavy metals, which makes it difficult to remove both at once. Therefore, before wastewater is released into the environment, it is ideal if the materials employed for treatment can remove both dyes and heavy metals [189].

So, in an attempt to address this environmental issue, many researchers have focused their efforts on enhancing current technologies or offering a substitute plan. Therefore their superior large surface area, high porosity, and customizable properties of their structures and functions

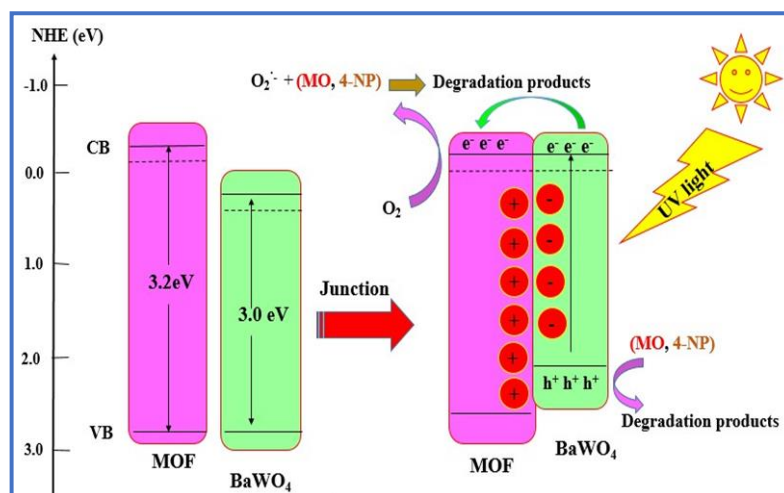


Fig. 14. suggested mechanism for the photocatalytic degradation of organic dyes (methyl orange and 4-nitrophenol) synthesized composite photocatalyst (MOF-199-NH₂/BaWO₄) under incident light irradiation. Reprinted from reference [192] Copyright (2023) with permission from Elsevier.

make metal-organic frameworks (MOFs) one of the appealing materials for this purpose [190]. For example Shi *et al.* synthesized Cu-MOFs/Fe₃O₄ as adsorbent for removal of lead (Pb(II)) and malachite green (MG) in wastewater. The synthesized Cu-MOFs/Fe₃O₄ composite was found to be the potential adsorbent for waste water treatment [191]. In the same year, Ramezanalizadeh et al. also reported the Immobilization of BaWO₄ nanostructures on a MOF-199-NH₂ an efficient photocatalyst for the degradation of methyl orange (MO) and 4-Nitrophenol (4-NP) water contaminants and Fig. 14 depicted the mechanism of methyl orange and 4-nitrophenol degradation using the synthesized composite. It's noteworthy to observe that the absorption maximum of the composites with worse optical properties than the pure MOF has redshifted as a result of the immobilization of BaWO₄ into the MOF-199-NH₂. Furthermore, the MOF-199-NH₂ has a bandgap of 3.2 eV, but the composite's predicted bandgap is lower at 3.0 eV (Fig. 14). Thus, for methyl orange and 4-nitrophenol, the MOF-199-NH₂/BaWO₄ compound allowed for full degradation in 50 and 80 minutes, respectively [192].

Sensing application

Different chemo-sensory materials are developed for sensing application but MOF-the composite sensor solves the drawback that is not solved by chemo-sensory materials due

to the properties of MOF-composite [193]. The property of MOF-composite includes a high porosity, a wide surface area, structural variety, flexibility, an exceptional capacity for adsorption, and chemical tenability [194]. The world's population increases from time to time because of environmental pollution and related health risks increase. Due to this materials for sensing application can be developed [195]. The development of effective analytical methods for the detection of biomolecules in clinical [196], environmental [197], and industrial applications [198] has a great potential for MOF-based sensors. MOF-composite is used to sense biological macromolecules like proteins and nucleic acids (DNA and RNA) [199] and small molecules including amino acids [200], Lipids [201], fatty acids [202], glycolipids, sterols, monosaccharides [203], phenolic compounds, and alkaloids [204]. To detect chemicals in different fields chemical sensors and photochemical sensors can be applied. Different gases were detected by MOF-composite sensor including acetone, CO₂, CS₂, n-propanol, isopropyl alcohol, H₂, pyridine, n-hexane, toluene, SO₂, methanol, H₂O₂, ethanol, formaldehyde, etc [205, 206]. The application of MOFs in a variety of sensing applications, including the pH sensor, the detection of pesticide residues, the clinical diagnosis of diabetes, and photothermal therapy, shows significant promise [207]. For food safety, MOFs detail every use of MOFs in food safety monitoring, including sample preparation,

Table 1. Application of different MOF composite materials for adsorption separation and sensing

MOF-composite	Application	Reference
[MPPyr][DCA]/MIL-101(Cr)	Separation of CO ₂ /N ₂	[209]
PSF-NH ₂ -MIL-125(Ti)	Separation of CO ₂ /CH ₄	[210]
Ag NP @Fe ₂ O ₃ @MOF	Separation of C ₂ H ₂ /CO ₂	[211]
MOF-2/SBA-15	Selective Adsorption separation of CH ₄ /N ₂	[212]
UTSA-100	Separation of C ₂ H ₂ /C ₂ H ₄ to purify C ₂ H ₂	[213]
ZIF-71	Separation of Olefin/Paraffin	[214]
UTSA-200	removal of trace C ₃ H ₄ from C ₃ H ₄ /C ₃ H ₆ mixtures	[215]
NKMOF-1-Ni	Separation of C ₃ H ₄ /C ₃ H ₄ /C ₃ H ₆	[216]
PAA@PVP@UiO-66-PA	Separation of H ₂ /CO ₂	[217]
TFP-TAPA-Bu	Adsorption separation of Xe/Kr	[218]
Ag@Ni-MOF74	Xe/ Kr Adsorption in a Xe selective	[219]
[Cu ₂ (L)(H ₂ O) ₂] _n mDMF·2nH ₂ O (FJI-Y11)	H ₂ /D ₂ separation	[220]
ZIF-67@NH ₂ -SiO ₂	Separation of hydrogen isotope H ₂ /D ₂	[221]
UTSA-16@CAU-10-H@γ-ALOOH	Adsorption separation of H ₂ /D ₂	[222]
TIFSIX-2-Ni-i	C ₂ H ₂ /C ₂ H ₄ and C ₂ H ₂ /CO ₂ separations	[223]
UiO-66-NH ₂ @CA	Adsorption removal of heavy metal ions in water	[224]
Fe ₃ O ₄ @TMU-32	High Capacity adsorption Hg(II) and Pb(II) Removal	[225]
UiO-66-GMA@ chitosan	Pb (II) adsorption	[226]
PCN-224/TA/PVDF	oil-water separation and organic dye adsorption	[227]
ZIF-67@PLA	Separation of oil-water	[228]
C ₃ N ₄ /Cu-MOF	Efficient separation of oil-water	[229]
Ce-BTC@RGO/GCE	Electrochemically detecting dichlorophen (sensor)	[230]
Cu-MOF-199/ SWCNTs/GCE	Sensing material for hydroquinone and catechol	[231]
MIL-101(Cr)-rGO	Detection of hydroquinone and catechol	[232]
PC-rGO/MOF-199/CPE	Sensing of Anticancer Drug Idarubicin	[233]
UiO-66-NH ₂ @TpTt-COF	Sensing of tetracycline	[234]
SiO ₂ CuOF-graphene-Pan	Effective Sensing of Ammonia	[235]
UiO-(OH) ₂ @RhB	Fluorescence Detection of Al ³⁺	[236]
Ni ₄ Mo/MoO ₂ @C	Sensing material for H ₂ S gas detection	[237]
JUK-8/CNP/PTFE/P	Detection of H ₂ and O ₂	[238]
Ni-Au /CNT/PVA	Detection of HIV DNA	[239]
Hemin@MOF-SA	Detection of H ₂ O ₂ and DNA	[240]
Cu@Co-MOF	Detection of glucose	[241]

separation, packaging, preservation, detection, and cleaning [208]. The MOF-composite and its adsorption separating materials and materials that can be detected by those sensors are listed briefly in Table 1.

CONCLUSION

The numerous research articles that have been published over the last two decades show how interesting MOFs are to both scientists and engineers. The interest in MOF-composites is due to their property including wide surface area, adjustable composition, highly flexible structures, and variable porosity. MOF-composite was made by metal ion/cluster with organic ligands with the

introduction of dopants like metal, metal oxide, carbon-based materials, enzymes, etc on its surface. The most common synthesizing techniques used to produce MOF-composite were encapsulation, Solvothermal, Solution impregnation, Click chemistry (reaction), and so on. Some examples of MOF-composite synthesizing by those techniques like MOF-199/N, Co-TCPP(Fe), NH₂-MIL-101(Al), DNA@ZIF-8, CA/ZIF-8@LAC/ MWCNTs/Au, etc. They are applicable for adsorption separation and as a sensor. MOF-composite is used to separate different gases like CO₂/N₂, CO₂/CH₄, C₂H₂/CO₂, C₂H₂/C₂H₄, C₂H₆/C₂H₄, C₂H₂/C₂H₆/CO₂/C₂H₄, C₃H₄/C₃H₆, C₃H₆/C₃H₈, noble gas separation and isotopic separation.

MOF-composite material widely applicable for separation of oil/water mixture, Separation of linear/branched alkane hydrocarbons (C₅-C₆-C₇), cyclic C₆ isomers (benzene/cyclohexane), cyclic C₈ isomers (styrene/ethyl benzene, and xylene isomers), Xylene isomers, and Styrene/ethyl benzene separation was done by MOF-composite. They are also used as sensors for sensing different toxic chemicals, drugs, gases, trace metals, contents in foods, etc. dichlorophen, hydroquinone, catechol, Drug Idarubicin, tetracycline, Ammonia, H₂S, H₂O₂, Al³⁺, glucose, etc. Generally, MOF composites are interesting materials with a wide area of application in many fields.

CONFLICTS OF INTEREST

The authors do not have any conflicts of interest.

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