



Review Recent Advances in Metal-Organic Framework (MOF) Asymmetric Membranes/Composites for Biomedical Applications

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Abstract: Metal-organic frameworks (MOFs) are a new class of porous crystalline materials composed of metal and organic material. MOFs have fascinating properties, such as fine tunability, large specific surface area, and high porosity. MOFs are widely used for environmental protection, biosensors, regenerative medicine, medical engineering, cell therapy, catalysts, and drug delivery. Recent studies have reported various significant properties of MOFs for biomedical applications, such as drug detection and delivery. In contrast, MOFs have limitations such as low stability and low specificity in binding to the target. MOF-based membranes improve the stability and specificity of conventional MOFs by increasing the surface area and developing the possibility of MOF-ligand binding, while conjugated membranes dramatically increase the area of active functional groups. This special property makes them attractive for drug and biosensor fabrication, as both the spreading and solubility components of the porosity can be changed. Asymmetric membranes are a structure with high potential in the biomedical field, due to the different characteristics on its two surfaces, the possibility of adjusting various properties such as the size of porosity, transfer rate and selectivity, and surface properties such as hydrophilicity and hydrophobicity. MOF assisted asymmetric membranes can provide a platform with different properties and characteristics in the biomedical field. The latest version of MOF materials/membranes has several potential applications, especially in medical engineering, cell therapy, drug delivery, and regenerative medicine, which will be discussed in this review, along with their advantages, disadvantages, and challenges.

Keywords: metal-organic frame works; MOF membrane; drug delivery; cancer therapy; asymmetric membranes

1. Introduction

All types of cancer and bacterial infections are serious threats to global health. According to reports until 2018, nearly 18 million people die annually due to cancer and bacterial infection [1]. The development of nanotechnology has experienced exponential growth in recent years and has led to entry into interdisciplinary branches of various sciences, such as biomedicine, bio-nano-technology, nanobiotechnology, and nanomedicine. Nanostructures



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Copyright: © 2023 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). are designed with acceptable biocompatibility and efficiency. This class of materials has attracted researchers and scientists to explore nanostructures as innovative and efficient nanoplatforms for anticancer drug/gene delivery and nanocarriers, as well as promising devices for various bioassay and diagnostic applications (especially for therapy/diagnosis of cancer) to be used [2-4]. Metal-organic frameworks (MOFs) are organic-inorganic hybrid materials that have been rapidly developed for various applications such as clean energy, storage of gases such as hydrogen and methane, catalytic processes, and drug delivery platforms [3,5]. MOFs are composed of organic ligands as bases and clusters of suitable metal ions as nodes. MOFs with different properties can be obtained by adjusting the almost infinite combination of metal nodes and organic ligands. The wide range in the choice of ingredients of MOFs makes it possible to adjust the different sizes of their pores from micropores to mesopores or macropores. Like other nanomaterials, one of the main characteristics of MOFs is their high surface area. In addition, by modifying the surface of MOFs, their performance increases. These properties make MOFs suitable candidates for biomedical applications such as drug delivery and magnetic resonance imaging (MRI) [6–8]. The high specific surface area and large pore size of MOFs facilitate the encapsulation of drugs with drug loads, and besides, the structural and functional flexibility of MOFs allows them to be adapted to shape size, and function [9,10]. Modifying the surface of MOFs with different chemical groups makes it possible to carry various drugs and can simultaneously act as MRI contrast agents. MOFs perform both diagnosis and treatment goals at the same time. According to the mentioned contents, MOFs offer new advantages in the field of biomedicine, including the fields of monitoring, diagnosis, and treatment [11,12]. The MOFs show greater than 90% porosity at physiological conditions, as well as a high degree of stability, whereas the MOFs have a high potential for biomolecular in situ functionalization with metals or organic molecules. Additionally, flexible membranes enable the design of bioactive substrates effectively. In addition, by using different metal groups and multiple organic molecules, structures with a tunable pore size (typically 0.4–6 nm) and specific surface area $(500-4500 \text{ m}^2/\text{g})$ of MOFs can be obtained [13–15]. ZIF-8 is one of the types of MOFs. In 2020 Ejeian, and colleagues [16,17], used polypropylene, polyurea, and ZIF-8 (PP/PDA/ZIF-8) as a selective thin film to support the essential activities of cells using dental pulp stem cells (DPSCs). The results of this study indicated that ZIF-8 nanostructures offer new opportunities for the surface functionalization of polypropylene membranes as nanomedicines with significant therapeutic benefits. The researchers tested the primary cell attachment, proliferation rate, and multilineage differentiation of cells on this platform. guided bone regeneration (GBR). In the continuation of this review, some applications of MOFs in the biomedical field and recent developments in this field have been discussed.

2. MOF Synthesis

In the still relatively young field of MOF synthesis, the chemistry of the solid state and zeolites have been recognized as forming the basis of this field [18]. As coordination polymers are formed by connecting metal ions with organic ligands as linkers, coordination chemistry has investigated compounds labeled as coordination polymers for years. Even though Prussian blue compounds and Hofmann clathrates showed reversible sorption properties, interest in porous coordination polymers emerged much later [19]. A layered co-trimester that demonstrated reversible sorption properties was first called MOF by Yaghi et al. around 1995 [20]. HKUST-1 and MOF-5, which were synthesized in 1999, became among the most studied MOFs in the following years [21,22].

Hydrothermal or solvothermal synthesis routines have traditionally been used to prepare MOFs on small scales through electrical heating, which can take several hours or days to complete. To prepare high-quality single crystals suitable for structural analysis in dilute liquid phase conditions, efforts were primarily focused on preparing high-quality single crystals. Later, microwave-assisted synthesis, sonochemical synthesis, electrochemical synthesis, and mechanochemical synthesis methods were used to shorten the synthesis time. Several studies were conducted on the scale-up of MOF synthesis conditions to obtain high yields of solid products for industrial applications; some of these studies also investigated the optimization of MOF synthesis conditions to achieve high yields [23,24]. A variety of elements, including zinc, copper, chromium, aluminum, and zirconium,

are commonly used to form frameworks containing bivalent or trivalent aromatic carboxylic acids or aromatics that contain N [25].

According to Table 1, the MOF structures were synthesized using the following methods and the key findings of these studies are presented.

Sample	Ligand	Metal	Synthesis Method	Ref
ZIF-8	2methyl imidazole	$Zn(NO_3)_2 \cdot 6H_2O$	Microwave-assisted	[26]
IRMOF-3	2-amino-1,4-benzene dicarboxylic acid	$Zn(NO_3)_2 \cdot 6H_2O$	Microwave-assisted	[27]
UiO-66	1,4-benzenedicarboxylic acid	$ZrCl_4$	Conventional solvothermal heating	[28]
Fe-MIL-100	1,3,5-benzenetricarboxylic acid	Metallic iron (Fe ⁰)	Conventional solvothermal heating	[29]
MOF-5	1,4-benzenedicarboxylic acid	$Zn(NO_3)_2 \cdot 4H_2O$	Conventional solvothermal heating	[30]
MOF-5	1,4-benzenedicarboxylic acid	$Zn(NO_3)_2 \cdot 4H_2O$	Microwave-assisted	[31]
ZIF-8	2methyl imidazole	ZnO	Mechanochemical synthesis	[32]
ZIF-4	Imidazole	ZnO	Mechanochemical synthesis	[32]
Al-MIL-100	1,3,5-benzenetricarboxylic acid	$Al(NO_3)_3 \cdot 9H_2O$	Electrochemical	[33]
Mg-MOF-74	2,5- 4 dihydroxy-1,4-benzenedicarboxylic acid	Mg(NO ₃)⋅6H ₂ O	Sonochemical	[34]
Cr-MIL-101	1,4-benzenedicarboxylic acid	$Cr(NO_2)_3 \cdot 9H_2O$	Microwave-assisted	[35]

Table 1. Some of the MOFs and synthesis methods.

3. MOF Membranes

MOF membranes are widely employed in the food industry, gas separation, drug delivery, and cancer therapy. various protocols have been applied for the design of MOF membranes. Thereby produced various types of MOF membranes with unique properties such as amorphous and low-crystalline MOF membranes, stimuli-responsive MOF membranes, ultra-thin MOF membranes, oriented MOF membranes, and pilot scale-up of MOF membranes. MOFs are gaining attention since they have regular nanopores and are highly selective in gas separation membranes. MOFs can be integrated into membrane materials in two different ways. In one way, MOF particles can be blended with a polymer matrix to form mixed matrix membranes (MMMs), while in another, dense MOF films can be grown on porous substrates. It is important in both cases to ensure the compatibility of MOFs with polymer matrixes and substrates at the interface [36,37].

3.1. Asymmetric Membranes

Asymmetric membranes were first manufactured out of cellulose acetate in the late 1950s by Loeb and Sourirajan and were used in reverse osmosis [38,39]. Since then, asymmetric membranes have been used in various fields of separation processes such as ultrafiltration, microfiltration, dialysis, gas separation, and wastewater treatment.

In addition to the mentioned applications, asymmetric membranes have also been used as wound dressings to treat skin injuries [40]. The first efforts of scientists in the field of regenerative medicine began with the development of occlusive wound dressings. Failure to absorb exudate in occlusive wound dressings such as Opsite[®], Omiderm[®], or Spandre[®] results in delayed healing in resuscitation medicine. Despite their effectiveness, these dressings did not prevent the penetration of microorganisms and dehydration of

wounds. Researchers later concluded that a combination of both systems (occlusive as well as macroporous) would be ideal, since the bacteria could not penetrate both structures and exudate could be absorbed and gas exchanged simultaneously [41].

The first wound dressings used in regenerative medicine were impermeable and prevented exudate from being absorbed through the wound, causing the healing process to be delayed. Eventually, microporous constructs (e.g., Coldex[®], Surfasoft[®]) were developed that allowed wound exudate to be better drained. Although these dressings prevented microbial penetration and wound dehydration, they were incapable of preventing penetration of microorganisms. The researchers concluded that combining both systems (occlusive and macroporous structures) would be the best option since it would prevent bacteria from entering while allowing exudate to be absorbed and gaseous exchange to occur. In order to improve blood flow, dressings were developed with a macroporous sublayer or a hydrogel linked to a dense or hydrophobic microporous top layer. These types of dressings include Lyofoam[®], Epigard[®], and Duoderm[®]. Although they were effective, they also had some disadvantages. For example, they had limited drainage capacity, exudate accumulation, and frequent replacements, which meant more wound infections. In the 1990s, Hinrichs et al., following Loeb and Sourirajan's work, developed for the first time an asymmetric polyurethane membrane (PU) [39]. Based on in vitro bacterial testing using P. aeruginosa, the asymmetric PU membrane exhibited an interconnected microporous top layer (pore size 0.7 m), which prevented wound dehydration and bacterial penetration. The sublayer also possesses a sponge-like structure that contains micropores (pore size: ten micrometers) and macropores (pore size: 50-100 µm) that enhance tissue regeneration and absorption capacities. Moreover, both layers acted as drug release reservoirs, allowing controlled gaseous exchange in a way that surpasses the limitations of Lyofoam[®], Epigard[®], and Duoderm[®] described in the previous paragraph. The use of chitosan (CS) membranes in asymmetrical forms has also been demonstrated. Due to its intrinsic properties, such as antimicrobial activity, biocompatibility, biodegradability and hemostatic properties, CS, a natural polymer derived from deacetylation of chitin, has been extensively used for wound dressing production. A platelet recognizes the CS surface and initiates coagulation in a matter of seconds by attracting the negatively charged residues on red blood cell membranes with its protonated amine groups, leading to a strong agglutination, thrombin production, and fibrin mesh synthesis within the microenvironment created by this polysaccharide [39,42]. Considering the various properties of MOFs, the use of these materials with asymmetric membranes have shown a high potential in separating different materials, especially gases, with the help of membranes [43]. However, the use of these two-dimensional materials and asymmetric membranes in the biomedical field, especially in the field of wound dressing, still has room for improvement.

3.2. Electrospinning

Normally, polymer melts or solutions are electrospun by either pressing them through a needle, coating them on a wire, rotating a cylinder, etc., or introducing them into a strong electric field. Generally, a strong electric field is used to melt or introduce the polymer solution [44,45]. Its higher production performance can be attributed to needleless technology, which can usually be scaled up from the laboratory to the industrial level. A biotechnology or medical application often uses nanofiber mats of this type [44,46]. Nanofiber mats are typically produced with incidentally oriented nanofibers using a static collector plate. Biotechnology and biomedicine often use these kinds of nanofiber mats. Nanofiber mats aligned in a particular direction are beneficial for cell growth [44,47,48]. A rapidly rotating collector and blades can be used to prepare them. A conductive substrate with grounding can be used in orienting nanofiber mats, including magnetic nanofibers, allowing fiber alignment. Dielectric coatings can also be applied to parts of the substrate to modify the electric field during electrospinning. In their study, Nguyen et al. study a cylinder collector shielded by two dielectric field, which controls fiber alignment and position through

diverting airflow due to cylinder rotation [49,50]. It is possible, as well, to create one or more layers with different mechanical properties by partially growing nanofiber layers on conductive structures and partly consolidating them. Different polymers and polymer blends can be used for electrospinning nanofiber mats. An electrospinning process can be carried out using aqueous solutions for some polymers. This method is simple and environmentally friendly. To use nanofiber mats in humid environments, a crosslinking step is required. The majority of water-resistant polymers must be electrospun using toxic or corrosive solvents, as opposed to electrospinning with low-toxic solvents such as dimethyl sulfoxide (DMSO). Electrospinning often uses polyacrylonitrile (PAN) due to its spinnability from DMSO. Another reason PAN is often electrospun is that it can be used as a precursor for carbon nanofibers. There is a wide range of materials that can be spun and co-spun into nanofiber mats, making them attractive for applications in many fields, including biotechnology and biomedicine. It is, however, for this discussion that we will be focusing on materials that are suitable for tissue engineering as well as other types of cell growth that apply to this discussion [51,52]. It is highly desirable for biomedical and biotechnological applications due to its low toxic nature and ability to be electrospun from dimethyl sulfoxide (DMSO). Its use for tissue engineering is less frequent, however than that of other polymers. Scientists found that electrospun nanofiber mats with low Fe-MOF levels were highly porous, had an appropriate fiber diameter, and were chemically stable [53,54]. Fe-MOF scaffolds showed better attachment, proliferation, and spreading of human umbilical vein endothelial cells (HUVEC) than pure PAN nanofiber mats, as shown in Figure 1. It was possible to exclude cytotoxic effects for Fe-MOF in low quantities; in vivo implantation did not result in any inflammatory response. The optimal concentration of Fe-MOF is the concentration that has a balance between scaffold degradation and an increase in cytotoxicity. A negative effect of Fe-MOF is that it negatively impacts cell activity, but it positively impacts pH values at the biointerface [54–56].



Figure 1. Various scaffolds after seeding HUVEC are shown in scanning electron micrographs. (a) Placing HUVEC on a PAN scaffold, (b) placing HUVEC on a PAN/5% FeMOF scaffold, (c) placing HUVEC on a PAN/10% FeMOF scaffold, and (d) placing HUVEC on a PAN/20% FeMOF scaffold. Reprinted from [54]. Copyright © 2021, MDPI.

4. Biomedical Applications

In coordination chemistry, open framework materials such as zeolites have been crucial since their discovery. In a wide range of applications, including catalysis, ion exchange, and gas separation, porous solids have been developed with targeted topology, architecture, crystallinity, and porosity [6,57,58]. Crystallized MOFs are open framework materials composed of metal nodes and organic ligands. The large surface area and well-defined pore distribution of MOFs have made them a valuable material for a wide range of applications since they were discovered at the end of the 1990s [59]. As well as tuning their chemical functionality during synthesis, they are capable of modifying their physical properties [60,61]. There has however been considerable effort made to improve the resistance of MOFs to degradation under different/harsh chemical conditions because of their chemical, thermal, and mechanical stability [62,63].

4.1. Biosensors

Biosensors allow for molecular interactions to be detected and converted into an electrical signal that can be detected [64,65]. An electrochemical, optical, piezoelectric, acoustic, or calorimetric biosensor involves the combination of biological elements, such as enzymes, DNA, RNA, metabolites, cells, and oligonucleotides [66–68]. An important characteristic of biosensors is their ability to convert biochemical interactions into electric signals that can be measured and quantified. Further, it is imperative that the minute changes that occur during biological processes, such as the interaction of various biomolecules, are analyzed efficiently. Therefore, biosensors are being developed to diagnose diseases, ensure food quality, and monitor the environment. As far as medical applications are concerned, biosensors are designed to detect tumors, bacteria, toxins, and biomarkers early in the onset of various disorders. Several factors have contributed to the popularity of biological sensors, including their low manufacturing costs, rapid response time, portability, high specificity, and high sensitivity, along with their ability to determine minute changes in biological elements [64,69–71]. Detecting biomolecules intracellularly presents a challenging task since non-specific targets can bind to biosensors [70,72]. To overcome this defect, structured materials from the Institute Lavoisier (MIL) family of metal-organic frameworks (MOF) are being utilized. In particular, a combination of MILs and fluorescence was used to detect intracellular ATP molecules. An indicator of active metabolism is the concentration of ATPs within the cell, which is the cellular energy currency [70,73]. As ATP aptamers (Apt-F) labeled with carboxyfluorescein (FAM) were linked to MIL-100, and thus quenched by PET (photoinduced electron transfer), Apt-F is bound to the ATP molecules at this point, thereby restoring FAM fluorescence. Using a chemiluminescent sensor, microfluidics-based nanosensors have been developed for identifying CD4 cells in the presence of HIV. Based on previous research, CD4+ and CD8+ T-helper cells are thought to play a role in wound healing, and so immunosensors can be used to monitor wound healing [64,74,75]. Due to the difficulty of finding water-soluble photosensitizer molecules that can bind selectively to tumor tissues that have a wide range of characteristics and are water-soluble (for high cell uptake), nanoscale agents are generally used for delivering photosensitizer molecules (molecules that destruct tumors upon exposure to light) [64,71]. With their high porosity, large surface area, and high tunability, MOFs can be used as nanocarriers for the treatment of sick tissue. The size, stability, half-life, and biocompatibility of these nano-carriers must also be monitored to increase their specificity to tumor tissue [76,77].

4.2. Drug Delivery

It has been proven that drug delivery systems (DDSs) consist of a carrier and a drug. Conventional DDSs such as tablets, capsules, and granules have several limitations and disadvantages such as high doses needed, poor bioavailability issues, and side effects. Due to various limitations of conventional DDSs, new DDSs have been created and designed in the last decade. Recently, numerous DDSs have been developed to reduce side effects and increase therapeutic efficacy. Therefore, inorganic materials such as carbon nanotubes, graphene, magnetic nanoparticles (NPs) of iron oxide, and gold nanoparticles were used. MOF membranes are a fascinating and versatile class of NPs, assembled from metal ions/clusters and organic linkers. MOF membranes have various properties, such as high porosity and surface area and special chemical and thermal stability. They have been applied in diverse applications, such as gas storage, photochemistry, catalysis, separation processes, adsorption properties, diagnostic, and antimicrobial properties, and delivery of a large variety of active drugs, biological gases, and cosmetics. Due to these properties, MOF membranes have become a good candidate for therapeutic and medicinal targets. The properties, such as low side effects, stimulus-based delivery systems, and multiple drugs loaded properties, have popularized the use of MOFs in drug delivery in the last decade. MOF membranes have been synthesized by diverse methods such as solvothermal, sonochemical, mechanochemical, and electrochemical. Drug release is controlled by MOFs, which have a high loading capacity, and targeting capabilities thanks to their high specific surface area, adjustable structure, modifications, and biodegradability [78]. Their physicochemical properties change as a result of being absorbed by proteins and cellular components [79,80]. This makes it extremely problematic for nanomaterials to have low therapeutic efficacy, poor targeting, and adverse side effects, which are all the result of their nano size. It is still possible to modify biomaterials (PEG, polymers, liposomes, hyaluronic acid, proteins, peptides, etc.) with MOF surfaces to some extent, despite MOFs' properties of biocompatibility, active/passive targeting, and long circulation. However, there are still some limitations, such as easy degradation, immune recognition, and low targeting efficiency [81–85]. Moreover, the immune system can easily recognize and eliminate synthetic composites as foreign materials. Hence, most synthetic MOF carriers have a short half-life, which makes it difficult for them to accumulate in diseased parts of the body [86,87]. The ideal drug carrier should therefore be biocompatible, stable, and able to target drugs. Physiochemically and biologically, biomimetic nanomaterials are derived from cell membranes [88–90]. Thus, they can avoid detection and elimination by the immune system as well as perform functions such as long circulation, targeted delivery, and controlled release. To achieve bionic camouflage, cell membrane biomimetic technology entraps MOFs and transfers their natural properties to the surface of MOFs using cell membrane biomimetic technology. Multifunctional MOF carriers can be designed easily using this method [91,92]. Since cell membrane-cloaked MOFs have similar properties to membrane-extracted source cells, they are capable of long circulation and disease-related targeting. Nano-carriers are currently made from various plasma membranes and cancer cell membranes, including those from blood-circulating cells [93,94]. The MOF platform has already been used to modify red blood cells (RBCs), platelets, neutrophils, macrophages, dendritic cells, and various cancer cell membranes to achieve biological camouflage [95–98]. The combination of enhanced bio-functions with improved drug delivery properties can be exhibited in a biomimetic MOF carrier. Considering the mentioned advantages of MOFs, the synergy of these properties with the capabilities that membranes bring can be considered a promising option in the field of controlled drug delivery [99]. Researchers are investigating porous polymers for applications apart from commercial applications, such as in catalysis and photo energy conversion [46,100]. For porous polymeric membranes, the most commercial potential may be found in the area of biomedicine, particularly in drug delivery, tissue engineering, bio-separation, and hemodialysis. Biodegradability and easy dissolution in certain solvents make these materials ideal for biomedical applications [85,101,102]. Biological scaffolds fabricated in vivo from membranes require controlled biodegradability, for example, for tissue engineering [103,104]. The development of porous polymeric membranes with diverse designs was enabled by selective solubility in polymer fabrication techniques such as electrospinning or block copolymer self-assembly [105]. The second half of this review discusses the use of porous polymeric membranes in biomedical applications. Detailed discussions of membrane properties, and features, including pore diameter ranges, porosity, and mechanical strength, will guide drug delivery techniques, including tissue engineering, biosensing, and bio-separation [87,106].

4.2.1. Biomaterial Types

Tissue engineering uses synthetic biodegradable polymers and acellular tissue matrixes such as decellularized tissues and organs [107,108]. These include natural polymers (such as collagen and alginate), acellular tissue matrixes, and synthetic biodegradable polymers (such as poly (glycolic acid) (PGA), poly (lactic acid) (PLA), and their copolymers) [109,110]. A biochemical, biomechanical, and biological evaluation of these biomaterials has been conducted. There are several differences between natural polymers and synthetic polymers, including their mechanical strength, degradation rate, and reproducible microstructures [46,111]. By electrospinning, nanofibers with unique properties such as high porosity, high permeability, and large surface area can be produced from natural and synthetic polymers. According to previous studies, a composite made of polycaprolactone (PCL) and collagen combines the excellent mechanical properties of PCL with excellent biocompatibility of collagen, making the composite a tissue engineering substrate for nerves, skin, bones, vascular scaffolds, etc. As a result, the electrospinning of PCL/Collagen can serve as an effective tissue-engineered substrate [112,113]. Since 2D nanomaterials possess unique physical, electronic, and chemical characteristics, they tend to possess remarkable properties compared to 1D and 3D nanomaterials [114]. In addition to having outstanding physical and chemical properties, 2D nanomaterials are also very biocompatible and biodegradable, which means they can be used in tissue or organ construction by combining them with cell biology [115,116]. Furthermore, 2D nanomaterials have high aspect ratios (surface area to volume ratios), which makes them suitable for use in drug delivery systems, as they can encapsulate a large number of molecules and provide superior control over the release of those molecules [117]. A variety of methods can be used to prepare MOFs, including solvochemical, electrochemical, mechanochemical, and sonochemical methods [23,77,118]. MOFs have proved a promising platform for biomedical applications due to their unique structure, large surface area, tunable pore sizes, and biodegradability. Rather than bridging metal ions with organic ligands, MOFs consist of organic bonds [119,120]. A polymer fibrous mat can have increased porosity by adding MOFs as high-porosity compounds. There are many composites of these porous materials reported that consist of fibers or monoliths of MOFs embedded in a polymer matrix [121,122].

4.2.2. Osmotic and Diffusion-Controlled Membranes

Osmotic-Controlled Membranes

A porous polymeric membrane diffuses drug molecules through its pores. The properties of the membrane are adjusted to control the permeation and release of drug molecules. As far as membrane systems go, there are two main types: (1) membrane systems controlled by osmosis, and (2) membrane systems controlled by diffusion [123,124]. A polymer membrane reservoir is usually used for osmotic systems, which are capable of permeating water but not drugs. This approach involves pore-forming agents being added to the membranes to create pores in situ. Meanwhile, diffusion-controlled membranes transport drugs across the membrane via drug diffusion, swelling of polymers, and degradation of the polymers [125,126]. Porous polymeric membranes are critical for the delivery of drugs because they must remain chemically and mechanically stable till the drug cargo reaches the target point, they must be uniform in size to control drug permeability and they should be easily adjustable to fit the drug's size. Block copolymer self-assembly methods have gained popularity for drug delivery applications due to their ability to control pore sizes in the nanometer range and low fabrication costs [126–128].

Diffusion-Controlled Membranes

Compared to intravenous and oral drug delivery methods, transdermal drug delivery generally offers several advantages. In contrast to intravenous and oral DDS that require the internalization of membranes, the membranes do not need to be internalized in the body, thereby removing many barriers, such as controlled drug release or discomfort from needles [129–131]. It can be achieved by either injecting the drugs directly into the stratum corneum (SC) or by injecting them directly into the hair follicles and sweat ducts. To prevent drugs from leaking out of the transdermal drug delivery membrane, a drug impermeable layer is placed on top of it. In the case of porous polymer matrixes, an adhesive layer is used to attach the polymer matrix to the skin, followed by a drug reservoir and porous membrane to control drug release [132,133]. It has been demonstrated that a wide range of polymers can be used to produce transdermal drug delivery membranes, including gelatin, semi-synthetic polymers like hydroxypropyl cellulose, nitrocellulose, or cellulose, as well as synthetic polymers such as polysiloxane, polyisoprene, polyester, polyurethane, polyethylene vinyl acetate, polyacrylamide, and PVA [134,135]. Using a low current, the charged drug molecules pass through the body safely without causing physiological damage. Iontophoresis is a commonly used technique for this system that has been widely studied. Artificial membranes made of biocompatible materials are the main component of almost all iontophoresis patches. A significant improvement in drug penetration is possible due to active transport because: (1) without releasing the drug into the bloodstream, the superficial distribution of the drugs can be controlled; and (2) rather than releasing drugs into the bloodstream, the superficial distribution of the drugs can be controlled. The versatility of MOFs, their porosity, their large surface areas, and their high drug uptake have made them popular as DDSs in recent years [134,136]. The bioactive molecules can be incorporated into MOF materials in different ways. A one-pot synthesis and loading method takes advantage of the coordinative properties of the bioactive molecule by soaking the MOF system in a saturated solution of the bioactive molecule. The limitations of these methods are related to poor solubility of the bioactive molecules, competition with solvent molecules, or the absence of coordinative properties of the molecules. Mechanochemical methods are an alternative strategy. An environmentally friendly method for synthesizing MOFs and immobilizing catalysts has been reported based on mechanochemical synthesis [137,138].

4.3. MOFs for Cardio-Vascular Implants

The prevalence of coronary artery disease (CAD) in industrialized societies is approximately 4%. The traditional treatment for artery occlusions is the administration of anti-thrombotic medications and small molecular weight nitric oxide donors [139]. The artery may also be blocked permanently by implanting a drug-eluting stent. The use of copper-based MOFs in antithrombotic coatings for cardio-vascular implants has recently become more prevalent. Using Cu-BTC as a catalyst, it has been established that bloodborne s-nitroso-cysteine can be converted into cysteine and nitric oxide in vitro [140,141]. S-Ntirosoglutathione can also be catalyzed by more complex Cu-MOFs. MOF/polymer composite materials show promise as lead materials for novel implants due to their nitric oxide release properties. In addition to these preliminary studies, Cu-BTC was directly grown on stent surfaces and demonstrated acceptable hemocompatibility in vitro. The MIL-101 (Fe) polycaprolactone composite, which was evaluated as an effective non-copper-based MOF for cardiovascular implants, used the MOF as mechanical reinforcement, a sustained drug release vehicle, as well as an MRI contrast agent. The interactions between MOFs and cardiomyocytes are also poorly studied [140,142]. A coating of Cu-BTC combined with polydopamine demonstrated effective surface modification of cardiovascular stents in vivo. The reduction of protein absorption, thrombus formation, platelet adhesion, and increased vasodilation, especially with nitric oxide donors that require catalytic degradation, resulted in a significant reduction in protein absorption, thrombus formation, and platelet adhesion (Figure 2). Other studies have reported that 3-(1H-tetrazole-5-yl) isophthalic acid-based MOFs reduced sympathetic excitability and prevented arrhythmia. In the future, MOFs will need to be compared to clinically used materials to determine their effectiveness and safety [120,140,143].



Figure 2. By continuously releasing earth-alkaline ions from MOFs, bone implants are improved, increasing bone regeneration and biomineralization (**A**). By enhancing osteoinduction and integration of relevant genes in the presence of specific MOFs (**B**), and by improving tissue integration and antibacterial effects (**C**), MOFs facilitate osteoinduction and integration. Reprinted from [140]. Copyright © 2021, Frontiers.

4.3.1. Tissue Engineering

Despite donor availability, the need for organ and tissue transplantation is still considered one of the biggest challenges in the medical field [144,145]. Tissue engineering scaffolds are materials that, by mimicking the extracellular matrix environment (ECM), enable connection to the cells of living tissues and can be implanted in the tissues of living organisms [146,147]. Tissue engineering scaffolds must have characteristics such as biocompatibility and high mechanical properties. On the other hand, the scaffold should have a suitable biodegradability speed for optimal tissue regeneration and match the new tissue regeneration speed [148–151]. The goal of regenerative medicine is to restore and maintain the normal function of injured or diseased tissues through the use of biological substitutes that mimic native tissues' anatomical and functional characteristics. Engineering, cell biology, and materials science principles are used in this approach [152,153]. Today, tissue engineering is largely based on cell-based approaches and scaffold-based approaches [154,155]. Traditionally, tissues are engineered using natural and synthetic scaffold materials. For proper direction and orientation of new tissue in growth, scaffold-based tissue engineering relies more on the body's natural ability to regenerate [53,156,157]. By mechanically and chemically removing cellular components from tissues, collagen-based tissue matrices can be prepared by constructing artificial microenvironments from natural or synthetic materials [158,159]. During cell ingrowth, extracellular matrix proteins replace scaffold materials that slowly degrade following implantation [160,161]. The field of tissue engineering, despite its challenges and versatility, has established itself as a very important discipline that combines alternative materials as replacements for damaged tissue and stimulates natural regeneration [155]. Modern science has made tissue engineering a dominant technique for overcoming transplantation limitations, graft rejections, and complexities in restoring function [162,163]. In addition to nanotechnology and biosensors, tissue engineering has expanded into the design of organs on chips, clinical trials on chips, microfluidics, genetic

manipulation, and more. Deciphering etiologies and repairing damaged tissues is possible using this technique [164,165]. With the growing clinical need to repair critical-sized bony defects, guided bone biomaterials have been developed for bone tissue engineering [54]. A range of treatment approaches can be used to achieve bone regeneration, including guided bone regeneration (GBR), especially in the maxillofacial region [166,167]. An osseous injury cannot be penetrated by rapidly growing fibroblasts because of a barrier between gingival connective tissue and alveolar bone tissue. Osteoblasts can selectively repopulate bone defects during healing by maintaining a secluded space during this process [16,168]. There are two main types of GBR membranes: non-resorbable and resorbable materials [16,169]. Collagen (Col) membranes are advantageous for biomedical applications since they are biocompatible and biodegradable, eliminating the need for a second surgery. Col membranes, however, exhibit unsatisfactory mechanical properties and rapidly degrade, making them unsuitable for medical applications [170,171]. A MOF has the potential to make a promising biomedical platform because of its unusual pore structure, large surface area, and ability to be functionalized in a variety of ways [172–174]. The solution to designing bioactive substrates is to immobilize enzyme MOFs onto porous and flexible membranes. A MOF is also suitable for combining short peptides, antibodies, and nucleic acids. MOF layers have therefore been successfully used to support cellular behavior in vitro as well as in vivo [140,173]. Multifunctional MOF-based biomaterials are being designed and synthesized at an accelerated pace. Due to its excellent chemical and thermal stability, pH sensitivity, and negligible cytotoxicity, zeolitic imidazolate framework-8 (ZIF-8) is an ideal candidate for use in bone regeneration processes [172] (Figure 3).



Figure 3. ZIF-8 base Polymeric bone regenerative platform. Reprinted from [172]. Copyright © 2021, Wiley.

4.3.2. Bone Adhesives

Bone adhesives with properties such as biocompatibility and prevention of shape changes in drugs used in the reconstruction of bone defects play an important role in bone graft repair surgery [175]. A multifunctional hydrogel containing catechol and chitosan modified with ZIF-8 nanoparticles (CA-CS/Z) was produced by Y. Liu et al. to stabilize graft environment, accelerate bone regeneration, promote osteogenic differentiation, and expand blood supply [176]. By chemically converting catechol and ZIF-8, they developed hydrogels with excellent wet adhesion. An analyses of the characterization data demonstrated that CA-CS/Z hydrogels were synthesized successfully, displaying reliable mechanical strength, excellent rheological properties, and excellent adhesion for clinical application. In rat bone marrow mesenchymal stem cells (rBMSCs), their superior biocompatibility allows them to provide blood supply through paracrine production of vascular endothelial growth factor (VEGF). Furthermore, ZIF-8 nanoparticles released from the hydrogels could enhance osteogenic differentiation of rBMSCs by up-regulating the secretion and production of collagen 1, osteocalcin, and alkaline phosphatase. Additionally, CA-CS/Z had antibacterial properties. CA-CS/Z also demonstrated an important effect on vascularized osteogenesis by stabilizing bone graft materials and thereby accelerating bone regeneration when applied to wound areas [175,176].

4.4. Cancer Therapy

Different synthetic methods have been used to prepare MOFs for a variety of uses, including catalysis, separations, sensing, drug delivery, optics, and many biomedical applications. Annually, millions of people die from cancer, one of the most serious threats to human health [177,178]. The most common method of treatment is chemotherapy. In addition to undesirable side effects, poor pharmacokinetics, and poor biodistribution, therapeutic drugs cannot be administered directly to patients. To decrease side effects and enhance therapeutic efficacy, research efforts have focused on controllable drug delivery vehicles [179]. Over the past few decades, various nanocarriers have been developed, including organic micelles, liposomes, and dendrimers, as well as inorganic mesoporous silica and quantum dots. These carriers, however, do have limitations in bio application, such as a low loading capability of liposomes, micelles, or dendrimers, and undesirable toxicity and degradation properties of inorganic materials. The following basic requirements must be met in a drug-loading system: a large loading capacity is important for intravenous drug administration; carriers must be nanoscale to allow intravenous drug administration; carriers must also be biocompatible, i.e., not toxic, and easily degraded by the body [179,180]. It is also possible to control the release of drugs from a vehicle loaded with drugs. Therefore, controlled drug release systems have been extensively investigated in cancer chemotherapy. A multimodal treatment system is also being developed to meet the ever-growing demand for effective therapeutic interventions [181]. Janus nanocomposites made of Au nanorods (NRs) and ZIF-8 were developed by Han and colleagues for CT imaging and synergistic chemo-photothermal treatment of BALB/c mice bearing the H22 gene (see Figure 4) [182,183]. MOFs have the following obvious advantages over traditional drug carriers, as discussed above: with a variety of morphologies, compositions, sizes, and chemical properties, MOFs have a variety of multi-functionalities and stimuli-responsive drug release mechanisms. In addition, MOF-based materials retain their desirable physicochemical characteristics after modification without significantly altering their controlled size, shape, or uniformity. The properties described above make MOFs beneficial as delivery systems for drugs, clinical tumor therapies, and other diseases [184]. Table 2 summarizes a few of the most important applications of MOF composites in biomedical fields.



Figure 4. LA-AuNR/ZIF-8 and its application for CT imaging-guided tumor treatment. Reprinted from [182]. Copyright © 2020, Wiley.

Table 2. MOF/Matrix applications in biomedical fields and preparation

MOF-Type	Matrix/MOF-Based Material	Preparation Method	Application	Ref
ZIF-11	Dimethylformamide	Electrospinning	Integrated restoration of the tendon-to-bone interface.	[106]
ZIF-8	Alumina membranes	Electrochemical deposition	Homologous-targeted therapeutic applications.	[185]
Cobalt-based MOF	PLA/PVP	Electrospinning	Antibacterial applications.	[186]
ZIF-8	Double-layer PCL/Col membrane	Electrospinning	Osteogenesis and angiogenesis applications. Bone regeneration, bone tissue engineering.	[172]
CuBTTri	Medical tubing	Tubing Coating Method/chemical vapor deposition (CVD)	Antibacterial coating for medical devices.	[187]
ZIF-8	PCL- PPCL- TAEG-PDA	melt-blown Electrospinning	Day ligDaylight recyclable antibacterial masks, medical protective clothing, and bactericidal air purification filter application.	[188]
ZIF-67	Au@Pt	One-pot synthesis ultrasound-assisted method	Electrochemical sensing of H_2O_2 in living cells	[189]
MOF-199,MOF- 74,MOF-5, MIL-53	Cotton	In situ cotton-MOF synthesis	Antimicrobial activity against nosocomial bacteria	[190]
MOF-5	Ag	Green and mild method	Antibacterial activity	[191]
Fe(III) metal-organic frameworks	PAN	Electrospinning	Tissue engineering investigations	[173]

5. Challenges

During the past decade, membrane surface engineering has become increasingly popular due to the use of mussel-inspired surface chemistry. The use of membranes in different processes and different membrane preparations have been extensively investigated and applied, but there are still several key challenges to be overcome in this field, which represent interesting future research opportunities. A common property of such coatings should be considered more closely. In membrane fabrication, mussel-inspired coatings may play a more diverse role due to their hydrophilicity. Multilayer composite membranes made from thin film composite membranes have shown excellent interfacial compatibility when they contain mussel-inspired interlayers. In this review, the latest applications of MOFs and membranes based on them in the biomedical field have been discussed. Because of their adaptability, excellent surface areas, high porosity, crystallinity, great loading capacities, improved thermal/chemical stability, and variable affinity, MOFs are most commonly used in biomedicine as multifunctional supports. Biomedical applications for MOF-based composites still face critical challenges despite significant advances in laboratory research. As a first step, toxicity issues regarding MOFs need to be addressed before or during clinical trials. The variety of MOF structures, species, sizes, and stabilities, along with the complex environment organisms live in, make MOFs toxic not only because of their composition, morphology, size, or stability, but also because of the environments they live in. As a result, a comprehensive assessment of MOF toxicity is necessary. To date, considerable research has been conducted on the toxicity of MOF scaffolds, with most experiments focusing on short-term in vivo or acute toxicity. However, the most important aspect of MOF scaffolds for biomedical applications is long-term toxicity, which has rarely been addressed. In vivo or acute toxicity studies of MOFs have been conducted in many excellent studies so far. Despite the critical aspect of long-term toxicity that must be considered if MOF scaffolds are to be outstanding biomedical candidates, few studies have been conducted to date. To evaluate MOFs' toxicity comprehensively, extensive in vivo studies and long-term tissue accumulation monitoring is urgently needed. A ligand with strong biocompatibility and metal ion nodes with high biocompatibility can also be used to create functional MOFs while avoiding toxicity associated with MOFs. Considering the potential properties of MOFs, their use in the construction of MOF-based membranes has become a promising option in the biomedical field. Because the synergism of the properties of MOFs with the characteristics of membranes that can be produced from various materials such as polymers can facilitate the controlled and stable release of drugs. In addition, the use of membranes based on tissue engineering has paved the way for many treatments. In recent years, advances in nanotechnology and the understanding of biosensors have allowed tissue engineering to be applied to designing organs on chips, conducting clinical trials on chips, using microfluidics, manipulating genetics, and so on, for the puerto decodend repairing drepairssues. Considering the variety of materials that can be used with MOFs, the field of application of these materials in biomedical field is even wider, while among these materials, there are plenty of materials that solve problems such as the low biocompatibility of MOFs. However, due to the novelty of this aspect of 2D materials, there are many undiscovered applications that need to be investigated.

6. Perspective and Conclusions

In this review, the latest applications of MOFs and membranes based on them in the biomedical field have been discussed. Because of their adaptability, excellent surface areas, high porosity, crystallinity, great loading capacities, improved thermal/chemical stability, and variable affinity, MOFs are most commonly used in biomedicine as multifunctional supports. Biomedical applications for MOF-based composites still face critical challenges despite significant advances in laboratory research. As a first step, toxicity issues regarding MOFs need to be addressed before or during clinical trials. The variety of MOF structures, species, sizes, and stabilities, along with the complex environment organisms live in, make MOFs toxic not only because of their composition, morphology, size, or stability, but also because of the environments they live in. As a result, a comprehensive assessment of MOF toxicity is necessary. To date, considerable research has been conducted on the toxicity. However, the most important aspect of MOF scaffolds for biomedical applications is long-term toxicity, which has rarely been addressed. In vivo or acute toxicity studies of

MOFs have been conducted in many excellent studies so far. Despite the critical aspect of long-term toxicity that must be considered if MOF scaffolds are to be outstanding biomedical candidates, few studies have been conducted to date. To evaluate MOFs' toxicity comprehensively, extensive in vivo studies and long-term tissue accumulation monitoring is urgently needed. A ligand with strong biocompatibility and metal ion nodes with high biocompatibility can also be used to create functional MOFs while avoiding toxicity associated with MOFs. Considering the potential properties of MOFs, their use in the construction of MOF-based membranes has become a promising option in the biomedical field. Because the synergism of the properties of MOFs with the characteristics of membranes that can be produced from various materials such as polymers can facilitate the controlled and stable release of drugs. In addition, the use of membranes based on tissue engineering has paved the way for many treatments. In recent years, advances in nanotechnology and the understanding of biosensors have allowed tissue engineering to be applied to designing organs on chips, conducting clinical trials on chips, using microfluidics, manipulating genetics, and so on, for the purpose of decoding etiology and repairing damaged tissues. Considering the variety of materials that can be used with MOFs, the field of application of these materials in biomedical field is even wider, while among these materials, there are plenty of materials that solve problems such as the low biocompatibility of MOFs. However, due to the novelty of this aspect of 2D materials, there are many undiscovered applications that need to be investigated.

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