



Nanoscaled metal-organic frameworks: charting a transformative path for cancer therapeutics and beyond

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Abstract

Through this inspirational note, we would like to highlight the potential of nanoscaled metal-organic frameworks within the biomedical field. The unique properties of these materials that make them promising candidates for new nanomedicines are assessed here as well as the progression reached so far for combinational cancer therapies and theranostic, along with its most recent advances in nanomedicine. Finally, the perspective and challenges of these materials within this field is discussed.

Keywords MOF · Nanocarriers · Combinational therapy · Cancer

Introduction

Metal-organic frameworks (MOFs) are a class of hybrid porous materials based on a coordinated network built from organic ligands and metal ions/clusters with potential voids [1]. Their exceptional porosity and versatility allows precise adjustments in composition and topology, resulting in fruitful tailored features for various fields (e.g. energy, environment, health) [2, 3]. Their recent miniaturization at the nanoscale level (nanoMOFs) has been a tremendous asset for their particular biomedical approach, since it strongly influences their administration and their *in vivo* fate. In other words, their physicochemical properties (e.g. particle size, surface charge, colloidal stability) will govern their affinity towards biological structures and/or matrixes, impacting also in their biodistribution, toxicity and, thus, their efficacy [4–6].

Compared to classical nanocarriers, nanoMOFs stand out as promising drug delivery systems (DDS) candidates due to several key parameters: (1) their chemical/structural versatility, enabling biocompatible material designs with potential control of their *in vivo* fate; (2) ideal adapted environment for encapsulating a wide range of active pharmaceutical ingredients (API: drugs, enzymes, nucleic acids, etc.), ensuring controlled releases under physiological conditions; (3) easy and potentially scalable synthesis,

following green methods with high yields; (4) proven *in vivo* safety (e.g. murine, nematodes, fish); (5) external surface engineering capability, endowing additional functionalities such as targeting, imaging, or enhanced stability in biorelevant environments [7–9].

In essence, despite its relatively short existence (less than two decades) in comparison with traditional nanomedicines, the MOF biomedical field has witnessed outstanding growth (Fig. 1) with remarkable progress in diverse therapeutic domains (e.g. cancer, bacterial/viral infections, vaccines) [5, 10–12]. Among them, antitumoral therapy stands out as one of the most advanced and demanded topic, achieving already the 1st clinical trial (phase I) in 2018 with a radio/immunodynamic Hf-based MOF for breast cancer [13]. Since then, alternative approaches such as immuno-, gene and/or photo-therapy have been developed over the time, pointing out a recent trend towards synergistic benefits arising from combinational therapy and/or theranostics [14–17].

Over the achieved MOF trajectory, P. Couvreur's involvement has been evidenced as a fundamental milestone, giving a unique and visionary perspective. Thanks to his expertise and guidance, he has significantly contributed to shaping the MOF biomedical path, fostering new insights. In a nutshell, Couvreur's contributions have inspired and empowered countless individuals to make their mark in this dynamic and rapidly evolving field, leaving a lasting legacy of progress and achievement.

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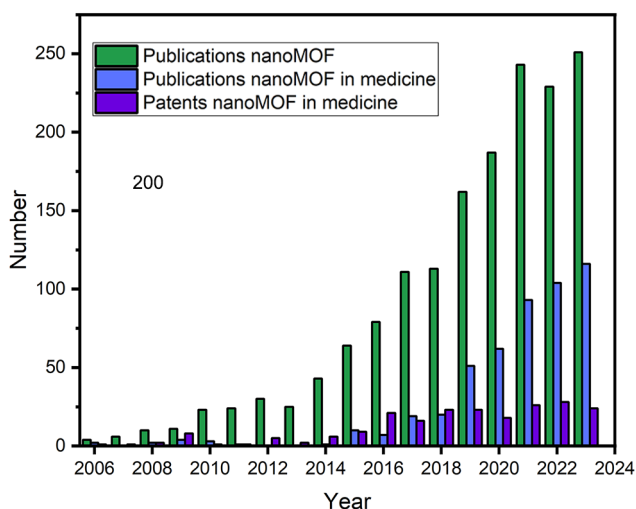


Fig. 1 Number of publications *per year* using as keywords ‘nanoMOF’, ‘nanoMOF in medicine’ and patents of ‘nanoMOF in medicine’. Publications were obtained from Web of Science and patents from Espacenet: (Jan 2024)

NanoMOFs origin in medicine: a pioneering journey

MOF exploration in DD [18] and imaging [19] started concomitantly in 2006. The pioneering MOF approach for medical imaging was proposed by Lin’s research group evidencing the gadolinium(III) nanoMOFs as potential contrast agents for magnetic resonance imaging (MRI), reaching exceptional longitudinal (r_1) and transversal (r_2) relaxivities [19]. In parallel, the incorporation of Eu^{3+} or Tb^{3+} enabled to obtain highly luminescent nanoMOFs as attractive multimodal imaging tool. In a step further, they demonstrated that high Mn^{2+} release in combination with a successful silica-coating lead to remarkable *in vitro* and *in vivo* r_1 potential; proving that the MOF stabilization by the surface anchoring moieties associated with an effective targeted delivery resulted in their enhanced antitumoral MRI performance [20].

At the same time, Férey/Serre’s group proposed for the first time DDS based on MOFs, providing the proof of concept of remarkable drug loading and controlled release abilities using two Cr(III)-carboxylate MOFs and the model drug ibuprofen. Despite the known Cr(IV) toxicity, this work emphasized the existence of isostructural non-toxic iron(III) carboxylates as a setting stage for safer DDSs [18]. Subsequently, the structural flexibility of some MOFs was established as an appealing asset for the monitorization of the MOF performance. Indeed, the flexible microporous Cr(III) or Fe(III) terephthalates (MIL-53(Cr, Fe)) were able to slowly release the ibuprofen with a totally predictable zero-order kinetics [21].

However, it was not until 2010 when the booming period of MOF in medicine truly started. Supported by an fruitful collaboration between Férey’s and Couvreur’s groups, Horcajada et al. reported a pioneer MOF nanopatform based on different iron(III)-carboxylate families combining DD and imaging performances (i.e. theranostic) [22]. The *in vivo* biocompatibility of these nanoMOFs was originally demonstrated, emerging as smart functional nanocarriers due to the efficient delivery of diverse challenging antitumoral/retroviral drugs as well as the high r_2 -relaxivity values associated to the Fe-based cores. In parallel studies, Taylor-Pashow et al. also demonstrated an interesting combined anticancer prodrug delivery with optical imaging of a Fe(III) aminoterephthalate MIL-101- NH_2 using human colon adenocarcinoma cells [23]. All these initial findings contributed to open new horizons for more precise tailored MOF nanomedicines.

NanoMOFs for next-generation therapeutic nanomedicine

The current clinical trend is the rational combination of therapies providing successful complementary/synergic effects. For instances, both molecular (biomarkers) and immune-targeted therapies have improved notably the outcomes over the past 2 decades [24]. In this scenario, MOFs have appeared as a promising nanotool for effective combined therapeutic performances. This new facilitating technology can be designed to tackle the demands of the most common administration routes (e.g. oral, *iv*, cutaneous) upon challenging illnesses (e.g. asthma, tuberculosis, COVID). Actually, the latest advances in MOF bioapplication have been focused mainly to the external functionalization, demonstrating their ability to effectively target a specific tissue [6]. A relevant example of this great potential has been the development of the 1st clinical trial of radio/immunodynamic Hf-MOF, RiMO-301 for cancer treatment [13]. These biomaterials were well tolerated, even with the immunotherapy pembrolizumab, with promising efficacy in patients with advanced tumors treated with concurrent palliative radiotherapy. The findings endorse the potential of RiMO-301 as radio-enhancer, prompting the initiation phase 2 investigations.

In this encouraging context, one of the most explored nanoMOF-based combined treatments has focused on reactive oxygen species (ROS)-based dynamic cancer therapies, including different external *stimuli* (e.g. photodynamic-PDT, sonodynamic-SDT, radiation dynamic-RDT, chemodynamic-CDT therapies) [25]. Thus, the general strategic trend seems to be based on combinational PDT, highlighting PDT/chemotherapy as the most emerging one [26–31]. If

chemotherapy is achieved with the drug entrapment within the MOF porosity, PDT is generally accomplished by using photosensitizers (i.e. as part of the frame or loaded in the cavities; such as porphyrins) and/or by different inorganic nanoparticles (NPs) association.

In contrast, combined SDT is a less explored approach with, however, attractive performances. For instance, studies involving a hypoxia-responsive CDT and SDT system based on a Cu(II)- nanoMOF with additional photoacoustic imaging properties; [32] and composites combining Pt or up-conversion NPs [33, 34] have been reported, achieving a synergistic *in vivo* therapeutic effect. Another promising combination is RDT with CDT. On this regard, a Hf- nanoMOF with a X-ray-triggerable prodrug (SN38) was developed, observing a synergistic RDT and chemotherapeutic effect, leading to an efficient tumor growth inhibition [35]. Although enhanced antitumoral performances have been proven, further research is needed to describe in deep their biosafety and biodistribution.

On the other hand, bearing in mind that cancer treatments have evolved from surgical resection towards (targeted) radio/chemo-therapy or immune-therapy approach [36], the nanoMOF immunological impact has started to be considered [31, 37–40]. A great illustration of this immune/chemoactive MOFs, reported by Lin's group [41], has been the Bi(III)-nanoMOF design for a dual radiotherapy and RDT combo in non-immunogenic tumors, achieving the macrophage repolarization and reduced immunosuppression. Thus, fostering the immune checkpoint blockade (ICB) which is one of the current first-line strategies against both primary tumors and metastases, suggesting a great potential for better antitumoral MOF efficacies.

Lastly and, following the latest tendencies of personalized medicines, great interest and milestones have been accomplished through gene therapeutics and DDSs, where nanoMOFs have been pointed out as effective gene nanovectors due to their cytoplasmic protection and release leading to relevant transfection performance. In fact, a promising gene and CDT approach has been reported using a Zn/Cu bimetallic nanoMOF loaded with rolling circle amplification substrates [42]. This system is activated by tumor-specific miR-21, generating DNAzyme for silencing EGR-1 mRNA with Zn²⁺ assistance, while a CDT is simultaneously induced through Cu²⁺ release. This claimed bioorthogonal strategy results in remarkable *in vivo* efficacy, demonstrating a high potential for tailored chemotherapeutics.

Another biomedical branch where nanoMOFs have played a meaningful role is in theranostics. Currently, pioneering methodologies are revolutionizing cancer diagnostics and treatment paradigms, mostly combining nanoMOFs with other NPs or specific molecules, allowing to monitor steadily the treatment response. In this sense, an efficient

NIR-triggered carbon dots / MIL-100(Fe) nanocomposite was reported for CDT-PTT along with fluorescence imaging (FI), permitting real-time tumoral growth tracking; [43] In another example, a polyethylene glycol (PEG) coated MnO₂-shrouded ZIF-8(Zn) was designed for targeted silenced RNA against crucial isozyme of triple negative breast cancer (TNBC), followed-up by MRI [44].

In essence, given the potential profitable MOF approach of combined therapies with multimodal imaging features, more sophisticated nanosystems have been recently addressed. Among the very nice reported examples [45–48], Bai et al. recently reported a multiple antitumor nanoreactor based on bimetallic Zn/Co-MOF, able to co-encapsulate metformin, cisplatin, indocyanine green, 1-methyl-tryptophan and glucose oxidase, being externally decorated with Mxene and AS1411-aptamer. This nanosystem demonstrated a synergistic PTT/PDT and immunological therapy in combination with trimodal imaging (MRI/FI/PTI) [49]. Despite their promising potential, these complex systems still require improvements to reach more effective clinical phases.

The road ahead for nanoMOFs

The strategic combination of nanoMOFs with smart NPs emerges as a novel perspective for creating highly effective theranostic systems. While *in vivo* preclinical studies have envisioned outstanding outcomes, it is imperative to strengthen efforts towards biosafe and biodistribution studies to finally move towards clinical trials. The transition from preclinical success to efficient clinical performance will be pivotal in realizing the full therapeutic capacity of these cutting-edge nanomedicines.

In this respect, the combination of targeted nanomedicines, including immune or gene therapy, with diverse treatment modalities holds the holy grail for advancing in any cancer treatment. It is essential to channel research efforts into this area in order to enhance the effectiveness of therapeutic strategies, clearing their path to productive clinical translation. In this sense, there is still plenty of room for improvement for a final nanoMOF formulation, capable to be fully exploited their capacity upon the administration routes and promote a faster transition to clinical.

While this emerging nanomedicine field has traditionally been predominantly fundamental, the rapid evolution towards more mature concepts, associated to extremely sophisticated and customized nanosystems, have provided promising cutting-edge therapeutic technologies. Nevertheless, the complexity of intricate systems might present challenges in terms of reproducibility, scalability and industry transferability. Although MOFs have been successfully

scaled up for other applications such as catalysis and gas storage, the scalability of this material in pharmaceuticals remains still a challenge.

Finally, balancing scientific innovation and practical applications is crucial for unlocking their full potential in clinics. In fact, the encouraging progress in recent years towards more mature and applied concepts is evident by the growing number of patents and the ongoing promising clinical trial of a nanoMOF-radio-enhanced therapy. Until now, the main focuses have been for antitumoral approach, so we encouraged the scientific community to dedicate efforts on the investigation of these promising multifunctional materials in other challenging diseases.

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Declarations

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