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Research Article

One Stone Two Birds: Zr-Fc Metal−Organic Framework Nanosheet for Synergistic Photothermal and Chemodynamic Cancer Therapy

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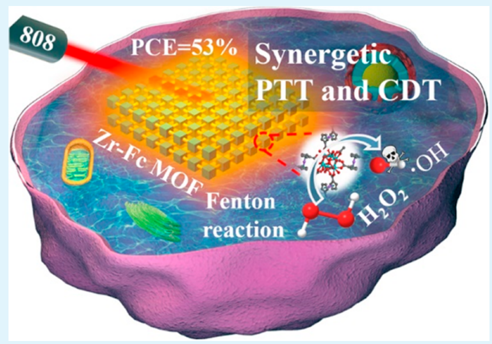
ABSTRACT: Metal−organic frameworks (MOFs) have been identified as promising materials for the delivery of therapeutics to cure cancer owing to their intrinsic porous structure. However, in a majority of cases, MOFs act as only a delivery cargo for anticancer drugs while little attention has been focused on the utilization of their intriguing physical and chemical properties for potential anticancer purposes. Herein for the first time, an ultrathin (16.4 nm thick) ferrocene-based MOF (Zr-Fc MOF) nanosheet has been synthesized for synergistic photothermal therapy (PTT) and Fenton reaction-based chemodynamic (CDT) therapy to cure cancer without additional drugs. The Zr-Fc MOF nanosheet acts not only as an excellent photothermal agent with a prominent photothermal conversion efficiency of 53% at 808 nm but also as an efficient Fenton catalyst to promote the conversion of H_2O_2 into hydroxyl radical (•OH). As a consequence, an excellent therapeutic performance has been achieved *in vitro* as well as *in vivo* through this combinational effect. This work aims to construct an "all-in-one" MOF nanoplatform for PTT and CDT treatments without incorporating any additional therapeutics, which may launch a new era in the investigation of MOF-based synergistic therapy platforms for cancer therapy.



KEYWORDS: *photothermal therapy, chemodynamic therapy, cancer therapy, two-dimensional material, metal−organic framework*

INTRODUCTION

Cancer is one of the main causes of death, which is responsible for approximately 9.6 million deaths in 2018 globally.[1] Although conventional therapies, for example, surgery, radiation therapy, and chemotherapy, have been adopted to cure cancer, the high recurrence, severe side effects, and multidrug resistance make them far from efficient therapies.[2,3] Owing to the intrinsic non-invasiveness and minimal side effects, photothermal therapy (PTT) has been considered to be a promising alternative approach by inducing localized hyperthermia using photothermal agents (PTAs), which convert light into thermal energy.[4,5] To achieve a highly efficient therapy, much effort has been spent exploring PTAs such as inorganic (gold and MoO_2),[6,7] organic (dye),[8] carbon-based (CNTs),[9] and polymeric (polyaniline and polypyrrole)[10,11] materials with a high photothermal conversion efficiency (PCE). In addition, the construction of a synergetic therapeutic platform by combining PTT with another treatment modality [e.g., chemodynamic therapy (CDT) and photodynamic therapy (PDT)] has also been identified as a promising strategy for obtaining a desirable therapeutic efficacy.

Metal−organic frameworks (MOFs) are porous nanomaterials with abundant micropores and large surface areas, which have been used for catalysis, gas storage, gas separation, etc.[12−14] Recently, MOFs have been used in PTT as a drug carrier for loading PTAs. Because of the highly porous structure of MOFs, various kinds of PTAs, such as Au nanorods,[15] indocyanine green (ICG),[16] polyaniline,[10] cyanine-containing polymer,[17] and polydopamine,[18] have been incorporated into MOFs for PTT. To achieve a better therapeutic effect, both PTAs and chemo agents[11,19,20] have been incorporated within a single MOF carrier to construct a synergetic platform. However, integration of two different components into a single carrier suffers from a complex preparation procedure. More importantly, in these cases, MOFs act as only a drug carrier by employing their porosity while other intriguing physical and chemical properties of MOFs are usually neglected.

In fact, MOFs have the potential to be an efficient PTA themselves considering the diversity of metal clusters and organic linkers.[21] By taking the advantage of the photothermal properties of MOFs themselves, researchers have found that it is much easier to construct a synergetic therapeutic platform (e.g., nitric oxide and PTT[22] or PDT and PTT[23,24]) by loading

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