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Synthesis and Characterization of pH-Sensitive Graphene Quantum Dot-Loaded Metal-Organic Frameworks for Targeted Drug Delivery and Fluorescent Imaging

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Abstract : Liver cancer is a significant global health issue, ranking fifth in incidence and second in mortality. Effective therapeutic strategies are urgently needed to combat this disease, particularly in regions with high prevalence. This study focuses on developing and characterizing fluorescent organometallic frameworks as distinct drug delivery carriers with potential applications in both the treatment and biological imaging of liver cancer. This work introduces two distinct organometallic frameworks: the cake-shaped GQD@NH_2-MIL-125 and the cross-shaped M8U6/FM8U6. The GQD@NH_2-MIL-125 framework is particularly noteworthy for its high fluorescence, making it an effective tool for biological imaging. X-ray diffraction (XRD) analysis revealed specific diffraction peaks at 6.81° (011), 9.76° (002), and 11.69° (121), with an additional significant peak at 26° (2θ), corresponding to the carbon material. Morphological analysis using Field Emission Scanning Electron Microscopy (FE-SEM), and Transmission Electron Microscopy (TEM) demonstrated that the framework has a front particle size of 680 nm and a side particle size of 55±5 nm. High-resolution TEM (HR-TEM) images confirmed the successful attachment of graphene quantum dots (GQDs) onto the NH2-MIL-125 framework. Fourier-Transform Infrared (FT-IR) spectroscopy identified crucial functional groups within the GQD@NH_2-MIL-125 structure, including O-Ti-O metal bonds within the 500 to 700 cm^{-1} range, and N-H and C-N bonds at 1,646 cm^{-1} and 1,164 cm^{-1}, respectively. BET isotherm analysis further revealed a specific surface area of 338.1 m^2/g and an average pore size of 46.86 nm. This framework also demonstrated UV-active properties, as identified by UV-visible light spectra, and its photoluminescence (PL) spectra showed an emission peak around 430 nm when excited at 350 nm, indicating its potential as a fluorescent drug delivery carrier. In parallel, the cross-shaped M8U6/FM8U6 frameworks were synthesized and characterized using X-ray diffraction, which identified distinct peaks at 2θ = 7.4 (111), 8.5 (200), 9.2 (002), 10.8 (002), 12.1 (220), 16.7 (103), and 17.1 (400). FE-SEM, HR-TEM, and TEM analyses revealed particle sizes of 350±50 nm for M8U6 and 200±50 nm for FM8U6. These frameworks, synthesized from terephthalic acid (H_2BDC), displayed notable vibrational bonds, such as C=O at 1,650 cm^{-1}, Fe-O in MIL-88 at 520 cm^{-1}, and Zr-O in UIO-66 at 482 cm^{-1}. BET analysis showed specific surface areas of 740.1 m^2/g with a pore size of 22.92 nm for M8U6 and 493.9 m^2/g with a pore size of 35.44 nm for FM8U6. Extended X-ray Absorption Fine Structure (EXAFS) spectra confirmed the stability of Ti-O bonds in the frameworks, with bond lengths of 2.026 Å for MIL-125, 1.962 Å for NH_2-MIL-125, and 1.817 Å for GQD@NH_2-MIL-125. These findings highlight the potential of these organometallic frameworks for enhanced liver cancer therapy through precise drug delivery and imaging, representing a significant advancement in nanomaterial applications in biomedical science.

Keywords : liver cancer cells, metal organic frameworks, Doxorubicin (DOX), drug release.
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