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# Hydrotalcite Monolayer toward High Performance Synergistic Dual-modal Imaging and Cancer Therapy

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## ABSTRACT

Recently, theranostic has drawn tremendous attention by virtue of the nanotechnology development and new material exploration. Herein, we reported a novel theranostic system by loading Au nanoclusters (AuNCs) and Chlorin e6 (photosensitizer, Ce6) onto the monolayer nanosheet surface of Gd-doped layered double hydroxide (Gd-LDH). The as-prepared Ce6&AuNCs/Gd-LDH exhibits a largely enhanced fluorescence quantum yield (QY) of 18.5% relative to pristine AuNCs (QY= 3.1%) as well as superior T<sub>1</sub> magnetic resonance imaging (MRI) performance ( $r_1 = 17.57 \text{ mM}^{-1}\text{s}^{-1}$ ) compared with commercial MRI contrast agent (Gd(III)-1,4,7,10-tetraazacyclododecane-1,4,7,10-tetraacetic acid (Gd-DOTA):  $r_1 \approx 3.4 \text{ mM}^{-1}\text{s}^{-1}$ ), resulting from a synergistic effect between AuNCs and Gd-LDH. In addition, both *in vitro* and *in vivo* therapeutic evaluations demonstrate an efficient dual-modality imaging guided anticancer performance, especially the synergistic enhanced magnetic resonance/fluorescence (MR/FL) visualization of tumor site. Therefore, this work demonstrates a successful paradigm for the design and preparation of LDHs monolayer-based theranostic material, which holds great promises in practical applications.

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## 1. Introduction

Theranostics, which combines diagnostic and therapeutic moieties into a single platform, can realize simultaneous diagnosis and therapy, real-time monitoring of drug distribution/delivery, and assessment of the treatment efficacy [1–6]. With the development of nanotechnology, a variety of inorganic and organic nanomaterials have been explored as theranostic agents with great potential applications in biomedicine [7–12]. Although much progress has been made [13–17], how to integrate diagnostic and therapeutic agent into one formulation, even with a largely

enhanced synergistic effect between each component, is vitally important for theranostics effectiveness but remains a challenge. Recently, ultrathin two-dimensional (2D) nanomaterials (*e.g.*, graphene, transition-metal dichalcogenides, hexagonal boron nitride, black phosphorus, etc.), have attracted considerable interest in theranostics, owing to their intriguing quantum size and surface property [18–21]. However, previous ultrathin nanomaterials synthesized via “top-down” mechanical-exfoliation strategy, show difficulties in a fine control over composition, size, thickness, and uniformity [22–24]. Therefore, developing a new drug formulation which affords imaging and therapeutic modality on the basis

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