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Iron oxide nanoparticle layer templated by polydopamine spheres: a novel scaffold toward hollow–mesoporous magnetic nanoreactors†

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Superparamagnetic iron oxide nanoparticle layers with high packing density and controlled thickness were *in situ* deposited on metal-affinity organic templates (polydopamine spheres), *via* one-pot thermal decomposition. The as synthesized hybrid structure served as a facile nano-scaffold toward hollow–mesoporous magnetic carriers, through surfactant-assisted silica encapsulation and its subsequent calcination. Confined but accessible gold nanoparticles were successfully incorporated into these carriers to form a recyclable catalyst, showing quick magnetic response and a large surface area (642.5 m² g^{−1}). Current nano-reactors exhibit excellent catalytic performance and high stability in reduction of 4-nitrophenol, together with convenient magnetic separability and good reusability. The integration of compact iron oxide nanoparticle layers with programmable polydopamine templates paves the way to fabricate magnetic-response hollow structures, with high permeability and multi-functionality.

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Introduction

Hollow and mesoporous structured nanomaterials exhibit a large surface area, unique permeability, high loading capacity and flexible functionalities, which has drawn tremendous attention in a broad range of technological fields such as catalysis,^{1,2} biomedicine^{3,4} and energy storage.^{5,6} Combining such nanocarriers with superparamagnetic iron oxide nanoparticles (SPIONs) would realize their unique magnetic operability and zero remanence/coercivity,⁷ which greatly favors their recycling or targeting (enrichment) abilities guided by external magnetic fields, when employed as heterogeneous catalysts^{8,9} or drug delivery vehicles.^{10,11}

Among the various strategies^{1,12} to create hollow–mesoporous nanostructures, the hard-templating method^{13–15} is quite straightforward and versatile, providing precise control over the size and uniformity of the final nanostructures. The incorporation of the desired functionalities could be achieved by modifying the template surface with various inorganic nanoparticles.¹² For SPION based magnetic nanocarriers,

achieving a compact shell of SPIONs on the template surface is rather critical, since the magnetic responsiveness of an individual single-domain sized SPION (smaller than 20 nm⁷) is generally weak in solution.¹⁶ For the conventional templates such as silica¹⁷ or polymer latex colloids,^{18,19} the major difficulties come from their native chemical inertness, restricting the facile deposition of compact functional shell materials. Moreover, the interparticle electrostatic repulsion becomes another obstacle in achieving high nanoparticle loading density on the template surface, in some aqueous-based assembly techniques.^{20–22} Strategies utilizing coordination,^{23–25} covalent bonding^{26,27} or electrostatic attraction^{28,29} to assemble SPIONs with solid templates have been established, to create secondary SPION structures with an improved magnetic response. Nevertheless, the assembly procedures are generally complicated, considering the multistep synthesis and surface modification of the templates and SPIONs, individually, together with a tedious layer-by-layer coating process^{24,25,28} when multilayered SPION nanostructures are desired.

The self-polymerization of dopamine is an emerging bio-inspired synthetic protocol for the creation of monodisperse polymer spheres and coating layers on virtually any surfaces.^{30–32} The advantage of taking polydopamine (PDA) as a template comes from not only its facile and well-controlled synthetic scheme, but also from its native highly reactive surface groups (catechols) for metal anchoring, compared with the commonly adopted polystyrene or silica templates. The strong binding affinity of catechol groups to iron oxide

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