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## **ACCEPTED MANUSCRIPT**

### Facile fabrication of gold particles decorated silk-on-silk nanocapsules

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#### **Abstract**

Both protein capsules and gold nanoparticles have attracted considerable attention in drug delivery systems. In this study, silk fibroin nanocapsules were fabricated by silk-on-silk layer-by-layer (LbL) assembly onto poly[lactic-co-(glycolic acid)] (PLGA) sacrificial templates followed by core removal. Subsequently, hybrid nanocapsules were fabricated by physical adsorption of DNA modified gold nanospheres and nanorods. These hybrid LbL-nanocapsules have potential applications for targeted drug delivery or combination therapy.

Key words: biomaterials; silk nanocapsules; gold; nanoparticles; layer-by-layer; hybridization

#### 1. Introduction

In drug delivery systems, polymeric vehicles with hollow interiors, especially layer-by-layer (LbL) capsules, have attracted enormous interest in recent years due to their distinct advantages for encapsulation, protection and delivery of active ingredients.[1-6] The original LbL technique is based on the simple alternative adsorption of oppositely charged polyelectrolytes on a sacrificial template via electrostatic interactions followed by core removal. The potential cytotoxicity arising from synthetic cationic components and time-consuming procedures of charge-compensated LbL-capsules stimulate the development of single-component capsules created by various synthetic polypeptides or natural biomacromolecules.[7-10] The shell thickness of polymeric capsules synthesized by one-step assembly can be regulated by adjusting molecular weight or the desolvated mass of shell composites.[8,9] Alternatively, microcapsules with controlled shell thickness were synthesized through hydrophobic interactions driven LbL assembly using silk fibroin, a biodegradable, biocompatible and excellent

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