



Contents lists available at ScienceDirect

Materials Letters

journal homepage: www.elsevier.com/locate/matlet

Gold nanocluster-coated gold nanorods for simultaneously enhanced photothermal performance and stability

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ARTICLE INFO

Keywords:

Gold nanorod
Gold cluster
Surfaces
Nanocomposites
Photothermal effect
Stability

ABSTRACT

Gold nanorods (GNRs) with unique optical properties are interesting nanomaterials, which have been widely explored in biomedical imaging and photothermal therapies. A GNR-based nanocomposite, which was coated with BSA-stabilized gold nanoclusters (GNCs), was successfully fabricated based on the electrostatic interaction between the negatively charged GNCs and the positively charged GNRs. Compared with the un-modified GNRs, this GNC–GNR nanocomposite with a core-satellite structure showed significantly enhanced photothermal performance together with improved shape stability and colloidal stability upon near-infrared irradiation. In addition, BSA as the stabilizer of GNCs can suppress the cytotoxicity of the nanocomposite, which made it well suited for further biological applications.

Nanomaterials with photothermal effect in near-infrared region (NIR) have attracted considerable attention for their potential biomedical applications [1,2]. Compared with other light source, NIR light can penetrate deeper into biological tissues, thus cause less damage to healthy tissues and cells. Among all the NIR-responsive nanomaterials, GNRs with tunable aspect ratios are of particular interest [3,4]. Application of GNRs needs to overcome a few obstacles, including cytotoxicity and stability. Although the cytotoxicity of GNRs can be successfully suppressed by methods such as protein coating [5] and PEGylation [6,7], low colloidal and shape stability often obstruct its application. Without sufficient protection, the heat generated from photothermal effect often melts GNRs into solid spheres, which eventually results in vanish of its photothermal performance [8]. Studies showed that photothermal stability of GNRs could be enhanced by embedding GNRs in a solid environment, such as carbon [9] and silica [10]. In addition, for potential biological applications of GNRs, it is crucial to minimize the dose to the patient while still providing sufficient amount of photothermal energy. Recently, we found that a coating layer of graphene oxide (GO) on GNR surface could enhance its photothermal performance and stability simultaneously [11]. Nanomaterials rather than GO need to be explored aiming at the improvement of overall performance of GNRs.

Gold clusters (GNCs) with high fluorescence have been highly attractive for its applications in bio-detection [12] and bioimaging [13]. The GNCs stabilized by bovine serum albumin (BSA) [14], which consists of 25 gold atoms (Au_{25}), are of particular interest due to the good biocompatibility of BSA. With excellent biocompatibility and

water-soluble nature, BSA has been widely utilized to suppress the cytotoxicity of nanoparticles [15] and to assist drug delivery [4,16].

Herein, we report on the surface modification of GNRs with BSA-stabilized GNCs, which provides not only good biocompatibility but also improved photothermal performance and stability to GNRs. We compared its photothermal performance and thermal stability with that of the un-modified GNRs under the same NIR irradiation (808 nm). The long-term stability of these nanomaterials was also investigated.

GNRs were synthesized via the seed-mediated growth method [3]. GNCs were synthesized by utilizing BSA as template and reductant [14]. GNC–GNRs were prepared by dropping as-synthesized GNC solution into GNRs solution, followed by incubation at 37 °C for 30 min. Fig. 1 shows the Transmission electron microscopy (TEM) images of GNC–GNRs in comparison with that of the as-synthesized GNRs. The average aspect ratio of GNRs was about 3.1, with a length of 30.7 ± 6.4 nm and a width of 10.2 ± 1.1 nm (Fig. 1A, Fig. S1). GNCs, which were synthesized via BSA-directed reduction, consisted of 25 gold atoms (Au_{25}) and were stabilized within BSA molecules [14]. As shown in Fig. 1B, the observed core-satellite structure confirmed the successful formation of GNC–GNRs assemblies. From the inserted high-resolution TEM image, it's clear that GNCs were directly attached to the GNR surface.

The as-synthesized GNRs contained two typical local surface plasmon resonances (LSPR) peaks, including a strong longitudinal LSPR (L-LSPR) at 719 nm and a weak transverse LSPR at 508 nm (Fig. 1C). After incorporation with GNCs, the L-LSPR peak displayed

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Received 24 August 2016; Received in revised form 31 October 2016; Accepted 13 November 2016

Available online xxxx

0167-577X/ © 2016 Published by Elsevier B.V.

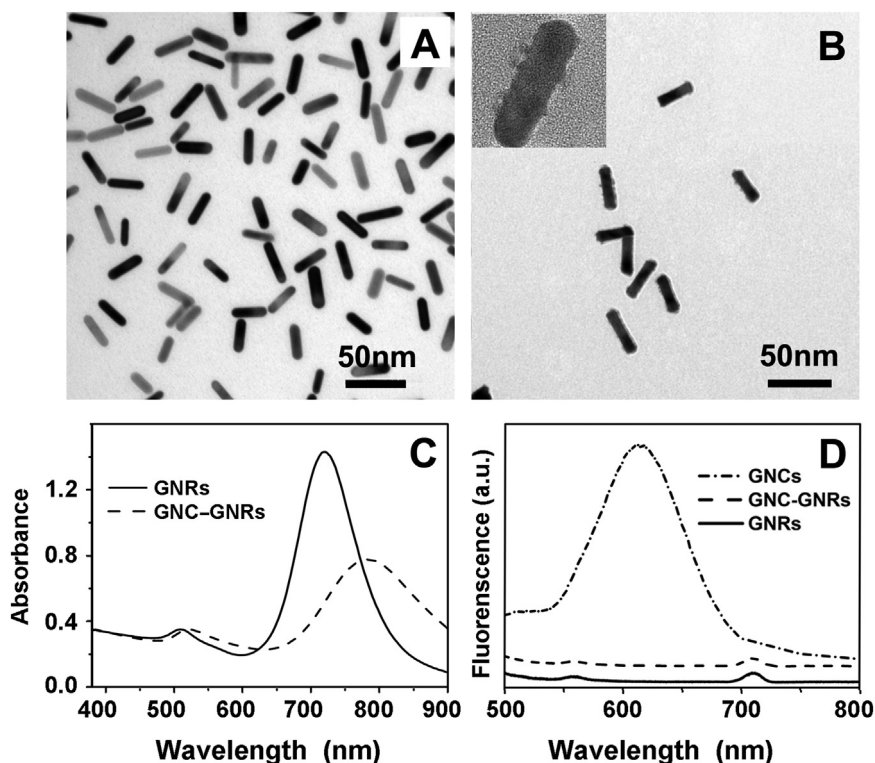


Fig. 1. TEM images of (A) GNRs and (B) GNC-GNRs. Insert is the Hi-resolution TEM image of GNC-GNRs. (C) UV-vis-NIR spectra of GNRs and GNC-GNRs. (D) Fluorescence spectra of GNCs, GNRs, and GNC-GNRs. Excitation wavelength was 470 nm.

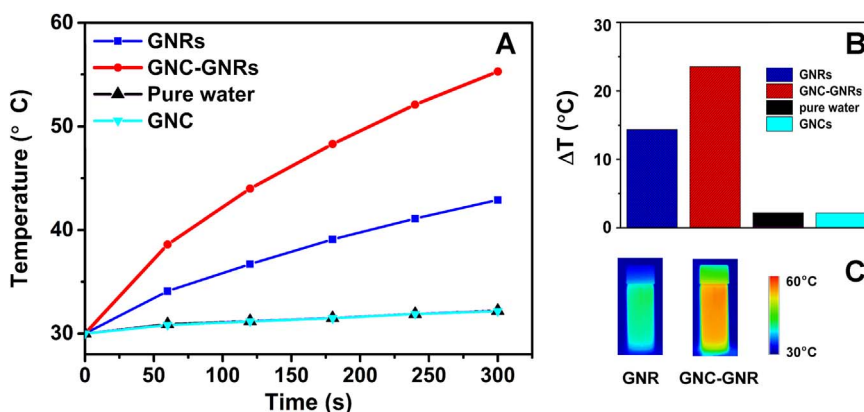


Fig. 2. (A) Thermal curves and (B) average temperature variations of water, GNCs, GNRs and GNC-GNRs upon NIR irradiation. (C) Corresponding thermo graphic images of GNRs and GNC-GNRs at the end of NIR irradiation.

an obvious red-shift from 719 nm to 786 nm. Similar red shift of the L-LSPR peak was detected for the BSA-coated GNRs which were fabricated under the same condition (Fig. S2). This considerable shift can be ascribed to a drastic change of dielectric constant surrounding the nanorods [17], which confirmed the formation of GNC-GNRs nano-assembly. Although with a peak broadening, the good shape of the L-LSPR peak provided strong evidence for mono-dispersion of GNRs upon GNCs modification.

Successful surface modification of GNRs was supported by Fluorescence spectroscopy and Zeta potential measurements. As shown in Fig. 1D, GNCs along exhibit a strong emission at 640 nm (excited at 470 nm). After incorporation with GNRs, this emission was completely quenched and only one small peak around 710 nm was observed, which should be the emission of GNRs [18]. No fluorescence recovery occurred when the GNC-GNRs solution was exposed to heating or NIR irradiation, indicating irreversible binding interaction between GNCs and GNRs. BSA-stabilized GNCs were negatively charged with a

zeta potential of -15.1 mV. GNRs displayed a positive zeta potential of $+31.5$ mV, which came from the positively charged CTAB bilayer packing on GNR surface. Therefore, formation of GNC-GNRs could be achieved based on the electrostatic attraction between the positively charged GNRs and the negatively charged BSA-stabilized GNCs. A corresponding decrease of zeta potential from $+31.5$ mV to -25.5 mV was observed for GNC-GNRs nanocomposite, confirming an efficient surface modification of GNRs.

Photothermal performances of GNRs and GNC-GNRs were investigated in solution state (OD=0.8). The concentrations of the nanorods in both solutions were approximately 0.2 nM [19]. Initial temperatures of all the sample solutions were set to be 28 °C. Each sample was exposed to the same NIR irradiation (808 nm, continuous wave, 2.96 W, 300 s) and the temperature change was recorded using a FLIR E40 thermal imaging system [20]. For both distilled water and GNCs, the temperature only increased by ~ 2 °C after NIR irradiation, indicating that they did not contribute much to the photothermal effect.

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