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Photothermally triggerable solid lipid nanoparticles containing gold nanospheres

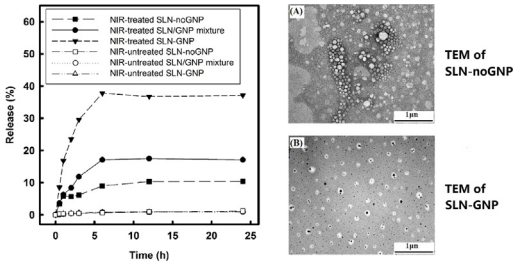
Juhyup Lee, Jin-Chul Kim*

College of Biomedical Science and Institute of Bioscience and Biotechnology, Kangwon National University, 192-1, Hyoja 2 dong, Chuncheon, Kangwon-do 200-701, Republic of Korea

HIGHLIGHTS

- Photothermally triggerable solid lipid nanoparticle (SLN) was prepared by including gold nanoparticle (GNP) in the matrix of SLN.
- The temperature of the SLN suspension of which GNP concentration was 6.6 μM increased from 26 to 36.6 °C upon 60 min-near infrared (NIR) irradiation, possibly due to the surface plasmon resonance oscillation of GNP.
- The NIR irradiation-induced release degree of Nile red (a fluorescence dye) loaded in the SLN was calculated to be as high as 38%.

GRAPHICAL ABSTRACT



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ABSTRACT

Photothermally triggerable solid lipid nanoparticle (SLN) was prepared by including gold nanoparticle (GNP) in the matrix of SLN. GNP was prepared by reducing gold ions in aqueous phase. The shape of GNP was spherical on TEM photo and the mean diameter, determined by dynamic light scattering, was 54 nm. GNP was dispersed in the melt of dodecanoic acid (DA) using cetyltrimethylammonium chloride as a stabilizer. The mixture melt (around 50 °C) was put in Tween 20 solution (0.5%, 1%, 2%) of the same temperature, emulsified and cooled in an ice bath to obtain SLN containing GNP. Thermograms revealed that Tween 20 solution (0.5%) allowed for the formation of SLN without an appreciable decrease in the melting point. GNP was found in the inside of SLN on TEM photo. The temperature of the SLN suspension of which GNP concentration was 6.6 μM increased from 26 °C to 36.6 °C upon 60 min-near infrared (NIR) irradiation, possibly due to the surface plasmon resonance oscillation of GNP. Despite the temperature during the NIR irradiation was far below the melting temperature of SLN matrix (around 43 °C), the NIR irradiation-induced release degree of Nile red (a fluorescence dye) loaded in the SLN was calculated to be as high as 38%. Since GNP was in close contact with the matrix of SLN, the fatty acid matrix adjacent to GNP could be locally heated, melted and fluidized, leading to a promoted release of the payload.

1. Introduction

Gold nanoparticles (GNP) have been proposed to be used as a photothermal agent for laser-based hyperthermia because they can convert light to heat efficiently [1–4]. When visible

* Corresponding author. Fax: +82 33 259 5645.
E-mail address: jinkim@kangwon.ac.kr (J.-C. Kim).

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light or near infrared light (NIR) is irradiated to gold nanoparticles, heat is generated due to the surface plasmon resonance (SPR) oscillation [5]. The wavelength where maximum SPR takes place depends on the shape of GNP (nanospheres, nanorods, nanoshells and nanocages). If GNP absorbs the light, electrons are exited and they collide with the lattice of gold. As a result, the lattice vibrates and produces phonons, and heat is generated by phonon–phonon interaction [6]. Light-triggerable liposome was prepared by incorporating GNP in dipalmitoylphosphatidylcholine/dimyristoylphosphatidylcholine liposome [7]. Upon UV light irradiation, heat is generated from GNP and it transfers to the liposomal membrane. Subsequently, the gel-to-liquid crystalline transition of the membrane takes place, leading to the triggered release of liposomal payload. Recently, GNP-loaded beads were developed by reducing gold ions captured in the bead matrix composed of alginate and poly(*N*-isopropylacrylamide-co-dimethyl aminoethyl methacrylate) (PNIPAM-DAEA) [8]. The bead exhibited NIR-responsive release in an aqueous phase because heat is generated from GNP by the light irradiation and it causes the phase transition of the thermo-sensitive polymer (PNIPAM-DAEA).

In this study, NIR irradiation-triggerable solid lipid nanoparticle (SLN) was prepared by incorporating GNP in the matrix of SLN. SLN is fatty acid-based spherical nanoparticle and it is prepared by melting fatty acid, emulsifying the melt in an aqueous phase, and solidifying the oil droplets upon quenching [9]. SLN has been extensively study in drug delivery system due to the low toxicity, the high bio-absorbability, the ease of preparation and the high loading efficiency of lipophilic drugs [10,11]. Since SLN is the crystalline particle of fatty acids, it becomes liquid oil droplet when the medium is heated to a temperature greater than the melting point of fatty acid. An active ingredient in SLN is almost immobile, but it will be relatively freely mobile in liquid oil droplet and readily will be released out of the droplet. In the present study, SLN was prepared by a melt-emulsification method using dodecanoic acid (DA) as a lipid. And gold microsphere was prepared by reducing gold ion in aqueous phase and it was used as GNP. For the hydrophobic modification of GNP surface, GNP was coated with cetyltrimethylammonium bromide (CTAB) though an electrostatic interaction between the surface and the head. Then, CTAB-coated GNP was incorporated in SLN by including it in the melt of DA and subsequently emulsifying the melt in hot aqueous phase. Nile red (NR) was also included in the melt as a lipophilic dye for NIR irradiation-triggered release experiment. The SLN containing GNP and NR was suspended in aqueous dimethylsulfoxide solution, and NIR was irradiated to the SLN suspension for 60 min. The release % of NR was calculated using definition, the percent of the amount of dye released with respect to the total amount of dye loaded. Fig. 1 shows the preparation of SLN containing GNP and NR, and NIR irradiation-triggered release of NR from the SLN. SLN containing GNP has never been reported before, and it can be used for the photothermal triggered release because the fatty acid matrix in contact with GNP could be locally heated, melted and fluidized, leading to a promoted release of its payload.

2. Materials and methods

2.1. Materials

Dodecanoic acid, cetyltrimethylammonium bromide (CTAB), trisodium citrate (dihy-drate), Nile red, Tween 20 were purchased from Sigma–Aldrich co. (St. Louis, Mo, USA).

Dodecanoic acid, Tween 20, CTAB, Nile red, gold, Hydrogen tetrachloroaurate (III) tetrahydrate was purchasedfrom Wako (Osaka, Japan). Water was doubly distilled in a Milli-Qwater purification system (Millipore Corp.) until the resistivity was18 MΩ/cm. All other reagents were in analytical grade.

2.2. Preparation of gold nanoparticles

Gold nanoparticle (GNP) was prepared by reducing gold ion in an aqueous phase [12,13]. Trisodium citrate was dissolved in distilled water so that the concentration was 270 mM. Chloroauric acid was dissolved in distilled water so that the concentration was 8.8 mM, then the solution was heated to 80 °C. In order to obtain GNP colloidal solution, trisodium citrate solution (1.25 ml) was added to the heated chloroauric acid solution (11.25 ml), then the reaction mixture was stirred for 10 min at the temperature of 80 °C. GNP suspension was cooled to room temperature (20 °C) and stored at 4 °C under dark condition.

2.3. Characterization of gold nanoparticles

GNP was characterized in terms of size, zeta potential, shape, photothermal conversion property [14–17]. The size distribution and the mean diameter of GNP were obtained by dynamic light scattering (DLS) technique. The light scattering intensity of GNP colloidal solution was adjusted to 50–200 count per second by diluting GNP colloidal solution with distilled water. The size was measured on a dynamic light scattering equipment (ZetaPlus 90, Brookhaven Instrument Co., USA). The shape of GNP was observed by transmission electron microscopy. GNP colloidal solution was transferred to a formvar/copper-coated grid and water was evaporated at room

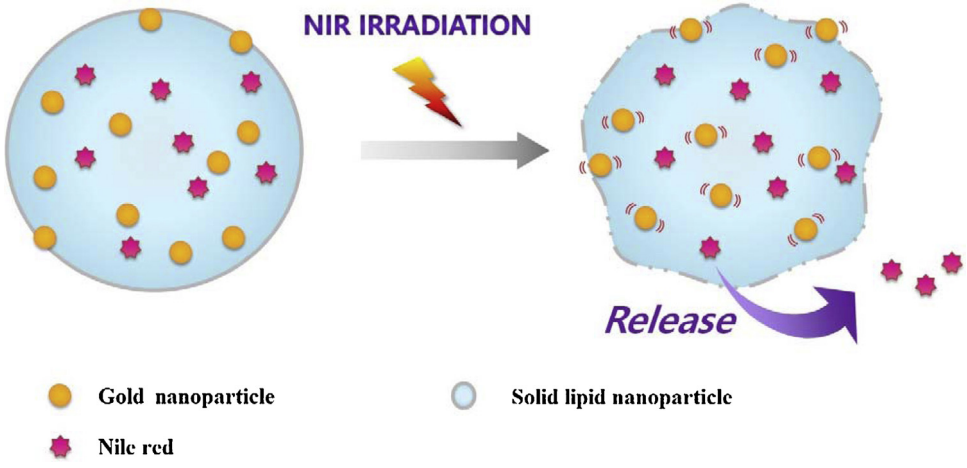

Fig. 1. Schematic diagram representing preparation of SLN–GNP and NR, and NIR irradiation-triggered release of NR from SLN–GNP.