



# ZnO-Au composite hierarchical particles dispersed oil-based nanofluids for direct absorption solar collectors

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## ABSTRACT

Direct absorption solar collector (DASC) is a promising technology for solar energy harvesting. Recently, nanofluids have shown great potential for DASC applications due to their tunable optical absorption properties. An advanced composite combining plasmonic gold nanoparticles (Au NPs) and hedgehog-like hierarchically structured ZnO particles (HPs) was prepared and used in DASCs. The nanofluid prepared with  $1.0 \text{ mg mL}^{-1}$  HP-Au exhibited excellent stability and broadband optical absorption properties, and reached an equilibrium temperature of  $\sim 125 \text{ }^\circ\text{C}$  within 1 h of solar heating at an irradiation power of  $10 \text{ kW m}^{-2}$ . A new photothermal conversion efficiency model was established to evaluate the solar harvesting performance of the HP-Au/oil nanofluid by simultaneously considering the effect of the working fluid temperature on the heating and cooling efficiencies. Compared to the base fluid, the HP-Au/oil nanofluid exhibits a 240% enhancement in photothermal conversion efficiency. The HP-Au/oil nanofluid shows great potential as a working fluid in direct absorption solar collectors.

## 1. Introduction

Nowadays, energy and the environment are two of the most important issues affecting society. With increasing energy demand and continual population growth, much research has focused on the development of new and sustainable energy [1]. Solar energy is regarded as one of the most promising candidates for sustainable energy in the near future owing to its non-polluting, abundant, and environmentally friendly nature [2,3]. Furthermore, the solar energy absorbed by the Earth's surface in 1 h exceeds mankind's energy consumption for an entire year. As a result, more and more attention is being directed to improving the utilization of solar energy [4,5]. Usual methods for the utilization and conversion of solar energy include solar-electrical, solar-thermal, and solar-chemical processes, among others [6]. The solar-thermal utilization is one of the most direct and straightforward approaches in which absorbed solar radiation is converted to heat for further use, such as power generation, water heating, and solar stills, and so on. Hence, the absorption of solar light and the photothermal conversion process are the key issues associated with the solar-thermal method [7,8].

In traditional solar-thermal methods, solar energy is absorbed by a black absorber surface. The heat is then transferred to heat transfer fluids through conduction and convection. However, there are usually

significant heat losses due to thermal radiation to the surroundings and thermal losses during heat transfer. As a consequence, direct absorption solar collectors (DASCs) that absorb solar energy directly through working fluids have been developed, in order to reduce these thermal losses.

Recently, nanoparticle suspensions (nanofluids) have shown great potential for DASC applications due to their tunable optical absorption properties [8,9]. In addition, nanofluids also have excellent thermo-physical properties that include high thermal conductivities [10,11], high thermal diffusivities [12–15], and high heat-transfer coefficients [16–19]. In 2009, Tyagi et al. [20] investigated theoretically the performance of a DASC containing an  $\text{Al}_2\text{O}_3$ -water nanofluid. Otanicar et al. [21] experimentally studied the effects of three kinds of nanofluid (carbon nanotubes, graphite, and silver-based) in a direct solar absorption system. Lie et al. [22,23] experimentally and numerically investigated the graphite or graphene based nanofluids for DASC and the results exhibit good solar harvesting performance. Yousefi et al. [24] experimentally explored the photothermal conversion efficiency of an  $\text{Al}_2\text{O}_3$ -water nanofluid and demonstrated a 28.3% enhancement in efficiency compared to pure water. Wang et al. [25] experimentally investigated the solar-collection performance of single-wall carbon-nanotube-based nanofluids for direct and volumetric solar steam generation. Chen et al. [26] investigated direct solar thermal energy

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harvesting with Fe<sub>3</sub>O<sub>4</sub>/silicone-oil nanofluids and found that thermal energy collection efficiency was enhanced by about 100% with the addition of homogeneously dispersed Fe<sub>3</sub>O<sub>4</sub> NPs. Even though nanofluids based on carbon materials exhibited good solar-energy absorption, the poor stabilities of these nanofluids seriously limit their potential applications.

In recent years, plasmonic nanoparticles have attracted much attention because of their localized surface plasmon properties and fast photothermal conversions. By using plasmonic nanoparticles in nanofluids, solar energy is efficiently converted into thermal energy. Chen et al. [27] experimentally studied the photothermal conversions of silver-nanoparticle-based nanofluids, resulting in improved solar energy conversion efficiencies. Wen et al. [28,29] investigated the photothermal conversion of gold nanoparticles and revealed the mechanism for high-efficiency solar-energy harvesting. Compared to the broad spectral range of solar radiation, there is usually a narrow absorption peak in the visible region for spherical plasmonic nanoparticles. Consequently, most of the solar energy is unable to be harvested by these nanoparticles. Some types of core-shell particles and blended plasmonic particles have been designed to realize broadband light absorption. Du and Tang [30,31] numerically studied the optical properties of plasmonic nanofluids containing different sizes, shapes, concentrations, and aspect ratios of gold nanoparticles for solar energy collection. Duan et al. [32] simulated the absorption properties of an Al/CdS nanoshell by using a finite difference time domain (FDTD) method and found that absorption enhancement was affected by coupling the core-shell interface, and the localized surface-plasmon effect. Li et al. [33,34] synthesized broadband Ag@TiO<sub>2</sub> core-shell nanoparticles in a one-pot method and achieved a solar steam-generation efficiency of 53.6% under an irradiance of 1 sun. Xuan et al. [35] experimentally and numerically investigated TiO<sub>2</sub>/Ag composite nanoparticles and demonstrated an excellent photothermal conversion efficiency. Lee and coworkers [36] numerically simulated the photothermal performance of a direct solar thermal collector containing SiO<sub>2</sub>@Au NPs. This study found that even with a low NP concentration (0.05 vol%), the plasmonic NP-based nanofluid achieved a solar collector efficiency of up to 70%.

Besides of the optical properties of nanoparticles, the dispersion of particles is also important for the using of nanofluids. Recently, a kind of hedgehog particles (HP, composite of polystyrene and ZnO) was proposed by Bahng et al. [37], which demonstrates unusual dispersion behavior in both hydrophilic and hydrophobic media. While ZnO only have absorption in ultraviolet region, which limits its application in solar thermal collections [38]. To enhance the solar light absorption, Au NPs could be used to decorate the ZnO and make use of spectral range in solar light. The photogenerated carriers in the Au NPs could enhance the light absorption and charge separation in the ZnO. Meanwhile, the coupling of ZnO and Au also allows tailoring of surface plasmon resonance (SPR) by electron transfer for plasmonic based chemical and biological sensors [39]. Furthermore, the hierarchical structure of HP may lead to multi-scattering effect during the light harvesting process, which is beneficial for the multiple absorption in NPs.

In this work, we hypothesized that plasmonic Au NPs composited with hedgehog-like hierarchically structured ZnO particles (HPs) are capable of realizing broadband light absorption. The composite particles (HP-Au) were prepared by a hydrothermal approach. To investigate their photothermal conversion performance, homogeneous nanofluids were prepared by dispersing the composite hierarchical particles in silicone oil. The light-response properties and stabilities of the HP-Au-based nanofluids were investigated. The photothermal conversion properties of these nanofluids were then investigated experimentally under solar light illumination. Finally, the solar photothermal conversion efficiency was calculated using a temperature-dependent model.

## 2. Experimental section

### 2.1. Materials

Zinc nitrate hexahydrate (Zn(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O, AR, 99%), hexamethylenetetramine (HMT, AR, ≥ 99.0%), zinc oxide nanoparticle (ZnO NP) dispersion (50 wt% in H<sub>2</sub>O), tetrachloroauric acid (HAuCl<sub>4</sub>, 49–50% Au), and sodium citrate dihydrate (HOC(COONa)(CH<sub>2</sub>COONa)<sub>2</sub>·2H<sub>2</sub>O, 99%) were purchased from Sigma-Aldrich and used as received. An aqueous suspension of polystyrene microspheres (μPSs) with a mean diameter of 1.00 μm and a concentration of 4.55 × 10<sup>10</sup> particles/mL were obtained from Polysciences, Inc., Warrington, PA, US (Catalog No. 08226-15). Silicone oil was obtained from Aladdin Reagent Co. Ltd., Shanghai, China. Double-deionized water (DDI water, Sartorius Water Purification System, arium® mini; 18.2 MΩ) was used in these experiments.

### 2.2. Synthesis of ZnO-Au composite hierarchical particles

Firstly, Au nanoparticles (NPs) were synthesized by the reduction of HAuCl<sub>4</sub> with sodium citrate [40]. A 950 mL aqueous solution of 0.458 mM HAuCl<sub>4</sub> was stirred vigorously and heated to boiling. Then 50 mL of an aqueous 34 mM sodium citrate dihydrate solution was added to the boiling HAuCl<sub>4</sub> solution, and boiling was continued for a further 20 min. The color of the mixture changed from yellow to colorless and finally to wine-red, which means that a well-dispersed solution of Au NPs had been prepared. The Au-NP dispersion was cooled to room temperature for further use.

A hydrothermal method was used to synthesize the hedgehog-like hierarchically structured ZnO particles (HPs) [37,41]. Firstly, 125 μL of the μPS solution was mixed with 4 mL of a diluted ZnO-NP dispersion (0.025 wt%) for 1 h to ensure that the ZnO NPs were completely adsorbed onto the μPSs. The ZnO-NP-coated μPSs were then dispersed in a 40 mL mixture of 25 mM Zn(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O and 25 mM HMT. Subsequently, a hydrothermal method was used to promote the growth of ZnO nanorods from the surfaces of the μPSs to form a hedgehog-like hierarchical structure.

The obtained HPs were then dispersed in 100 mL of the prepared Au-NP dispersion. Due to the electrostatic adherence between the ZnO NRs and Au NPs, the HPs become decorated with monodispersed Au NPs. Finally, the ZnO-Au-composite hierarchical particles (HP-Au) were collected as a powder using lyophilization and stored for further use. The schematic diagram for the preparation of the HP-Au particles is illustrated in Fig. 1.

### 2.3. Preparation of the oil-based nanofluids

A two-step method was used to prepare the HP-Au-dispersed silicone-oil-based nanofluids. Firstly, three different quantities of HP-Au powder were added into 20 mL aliquots of silicone oil and the mass concentrations of the nanofluids were adjusted to 0.1, 0.5, and 1.0 mg mL<sup>-1</sup>. The dispersions were ultrasonicated in a water bath for 10 min and magnetic stirred for 1 h. Finally, the homogeneous and stable HP-Au/oil nanofluids were obtained and stored for the photothermal conversion experiments.

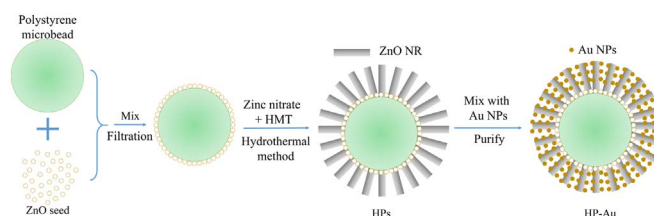


Fig. 1. Schematic diagram for the preparation of the HP-Au.

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