

## Volumetric solar heating and steam generation via gold nanofluids



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### HIGHLIGHTS

- Novel experiment was performed for nanofluids at a focused solar flux of 280 Suns.
- Strong surface evaporation was enabled while the bulk fluid was still subcooled.
- A new integration method was used to calculate photothermal conversion efficiency.
- Gold nanofluid (0.04 w%) increased photothermal conversion efficiency by 95%.
- No nanoparticle was entrained with steam even under vigorous boiling.

### ARTICLE INFO

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### ABSTRACT

Volumetric solar absorption using nanofluids can minimize the thermal loss by trapping the light inside the fluid volume. A strong surface boiling with the underneath fluid still subcooled could have many interesting applications, whose mechanism is however still under strong debate. This work advanced our understanding on volumetric fluid heating by performing a novel experiment under a unique uniform solar heating setup at 280 Suns, with a particular focus on the steam production phenomenon using gold nanofluids. To take the temperature distribution into account, a new integration method was used to calculate the sensible heating contribution. The results showed that the photothermal conversion efficiency was enhanced significantly by gold nanofluids. A three-stage heating scenario was identified and during the first stage, most of the energy was absorbed by the surface fluid, resulting in rapid vapor generation with the underneath fluid still subcooled. The condensed vapor analysis showed no nanoparticle escaping even under vigorous boiling conditions. Such results reveal that nanoparticle enabled volumetric solar heating could have many promising applications including clean water production in arid areas where abundant solar energy is available.

### 1. Introduction

Solar energy is the most dominant renewable source that is available and accessible to everyone, but facing many challenges to achieve efficient utilization [1]. Wide-spread solar powered applications are not limited to but consist of electricity generation [2,3], micro thermal power [4], chemical production line for methanol [5] and hydrogen [6], water desalination [7–10], greenhouse growth in agriculture [11], sterilization [12] and cooling and refrigeration [13,14]. The solar energy utilization of these applications can be significantly enhanced by suspending various nano-sized particles in a fluid, which is called direct absorption volumetric solar collectors [15–19]. In contrast to

conventional solar collectors [20,21] where the solar absorption is surface-based, i.e., having large radiative and thermal losses due to high surface temperature [22], the volumetric solar collectors minimize these losses by thermal trapping [23,24] and reduced temperature difference between the absorber and the fluid [25,26].

A variety of direct absorption nanoparticles have been analyzed in terms of the enhancement in the photothermal performance, including Ag [27–29], Au [30–32], CNT (carbon nanotubes) [33–35], Cu [36], Al<sub>2</sub>O<sub>3</sub> [37,38], graphite [17], graphene [22], and TiO<sub>2</sub> [39]. In addition to the volumetric heating, direct vapor generation due to localized heating of nanoparticles [40–43] is a recent development in this area. For example, Neumann et al. [44] showed that by using very dilute gold

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nanoparticles (16.7 ppm) under a focused solar light via a typical Fresnel lens, steam was produced instantly while the measured bulk temperature was still 6 °C approximately. The calculated steam generation efficiency reached 80%, meaning only 20% of the solar radiation was used to increase the bulk fluid temperature. Later simulation work [44–46] showed the possibility of nanobubble formation based on a non-equilibrium phase change assumption. However these results are quite different to the recent results from Jin et al. [47]. Still using a Fresnel lens (i.e. solar flux  $\sim 220$  Suns), it revealed that steam generation was mainly caused by localized boiling and evaporation in superheated regimes due to a highly non-uniform temperature distribution, albeit the bulk fluid was still subcooled. The hypothesized nanobubble, i.e., steam produced around heated particles, was unlikely to occur under normal solar radiations. It shall be noted that all these experiments [44,47,48] were performed outdoor, where the solar flux varied from time to time, and the focus by Fresnel lens limited the heating to a small area, leading to a non-uniform solar energy input. Such would lead to a very high solar flux in localized areas, producing spot heating and high evaporation rate locally.

As far as the steam generation mechanism is concerned, it has been shown analytically that a minimum radiation flux of  $3 \times 10^8$  W/m<sup>2</sup> is required to produce nanobubbles on heated nanoparticles [46,49,50], which can only be reached by powerful laser beams. In a separated study, Julien et al. [51] showed that  $1 \times 10^{10}$  W/m<sup>2</sup> was required to generate a nanobubble on a plasmonic gold nanoparticle. However quite differently, Hogan et al. [52] reported that  $\sim 1$  MW/m<sup>2</sup> solar reflux was sufficient for efficient steam production due to a collective effect of nanoparticles that both scatter and absorb light, hence localizing light energy into mesoscale volumes.

It shall be noted that most of the experiments performed so far were not under well-controlled conditions [44,47,48]. Beside the problem of varying solar flux and spot heating mentioned above, most of the experiments were performed by a single-point temperature measurement, ignoring the temperature distribution in the bulk fluid [32,44,53]. Though Jin et al. [41] and Ni et al. [43] used multipoint temperature measurement, only the average temperature was used for the evaluation of the photothermal efficiency. In Jin's work [41], the spot heating and small fluid volume minimized the temperature stratification phenomenon, and the fluid reached saturated boiling rapidly, where the most interesting phenomenon under subcooled condition was insufficiently captured. In addition, possible escaping phenomenon of nanoparticles with the steam under saturated boiling has not been investigated, which is critical for any potential desalination or clean water production applications. Clearly a better understanding of the solar steam generation by nanoparticles is much needed.

This work aims to advance the field by answering three questions: i) Would the steam generation phenomenon be different under a uniform solar heating, instead of spot heating? ii) What is the underneath mechanism for steam production if not by forming nanobubbles? and iii)

Would nanoparticle be escaped with the produced steam? To answer these questions, we performed a well-controlled experiment under a unique high power solar simulator (i.e. up to 4 MW/m<sup>2</sup>) with a large focus area to provide uniform heating. A novel one-dimension test section was designed, and multiple thermocouples were used to reveal the temperature distribution along the heating path. A novel integration method was proposed to calculate the sensible heating contribution and to aid the analysis of steam production mechanism. Various concentrations of gold nanofluids were produced and used as the test fluids, and the generated steam was condensed to reveal the presence of any nanoparticles. All sample nanofluids before and after the experiments were carefully characterized in terms of stability, size distribution and morphological variation. Such would allow us to answer the proposed questions and advance the solar applications by direct absorption nanofluids.

## 2. Materials and methods

### 2.1. Reagents and devices

Hydrogen tetrachloroauric acid (HAuCl<sub>4</sub>, Au  $\geq 49\%$ ) and Tri-sodium citrate (Na<sub>3</sub>C<sub>6</sub>H<sub>5</sub>O<sub>7</sub>, 99.8%) were purchased from Fisher Scientific and used as received. Deionized water was used throughout the experiments.

A transmission electron microscopy (TEM) (TECNAI, TF20) equipped with EDX (Energy Dispersive X-ray spectroscope), was used to analyze the morphology of the synthesized nanoparticles. The concentration of the gold dispersion was determined by an Atomic Absorption Spectrometer (AAS) (VARIAN, AA240FS). The hydrodynamic size and zeta potential of the nanofluids were obtained by a DLS (dynamic light scattering) device (Malvern nanosizer). The optical absorption of the nanofluid was examined by a UV/Vis spectrophotometer (HITACHI, U-3900) using a high precision cell with light path of 10 mm.

### 2.2. Nanofluid synthesis and characterization

Gold nanoparticles (GNPs) were synthesized by the one-step method based on a modified thermal citrate reduction method as reported by Zhang et al. [32] and Chen et al. [54]. In the synthesis process, 100 ml of 5 mM HAuCl<sub>4</sub> solution was mixed with 100 ml of 10 mM trisodium citrate solution. The resultant mixture was heated to the boiling point until the mixture turned to red wine color. The resultant solution was continuously heated at 80 °C in a sonication bath for further 3 h. Synthesized GNPs were aged at room temperature for 24 h and cleaned by dialysis from 8 kDa membrane. The membrane allows the excessive ions to diffuse smoothly from the suspension and blocks the GNPs. DI water was changed twice a day for a period of 10 days, leading to pure GNPs dispersions. The concentration of the resulting nanofluid was

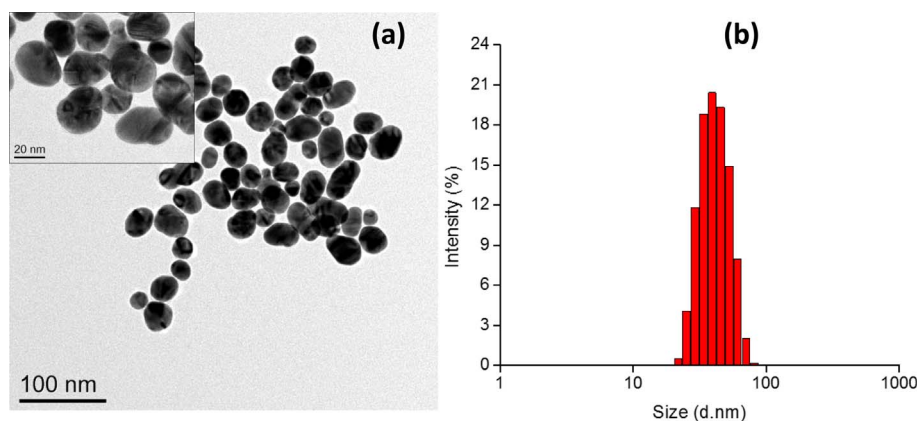


Fig. 1. Characterization of the synthesized gold nanoparticles, (a) TEM image of the gold nanoparticles showing a good suspension and size variation and (b) hydrodynamic size distribution of the gold nanoparticles measured by DLS.

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