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Investigation of photothermal heating enabled by plasmonic nanofluids for direct solar steam generation

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A B S T R A C T

Steam production has a wide range of applications such as seawater desalination, waste sterilization, and power generation. The utilization of solar energy for this purpose has attracted much attention due to its inexhaustibility and pollution-free nature. Here, direct solar steam generation at low-concentrated solar power using plasmonic nanofluids containing gold nanoparticles (Au NPs) was investigated experimentally. The key factors required for highly efficient solar steam generation, including Au NP concentration and solar power intensity, were studied in a simulated solar system by measuring the water weight loss and system temperature change. The best evaporation performance was obtained using a plasmonic nanofluid containing 178 ppm of Au NPs under 10 sun (1 sun = 1 kW m^{-2}) illumination intensity, and the total efficiency reached 65%. However, the total efficiency of pure water was only 16%, which means that the plasmonic nanofluids reached a \sim 300% enhancement in efficiency. Higher solar power led to a higher evaporation rate, higher specific vapor productivity (SVP), and higher Au NP concentrations resulted in better evaporation performance. Localized solar heating at the fluid-air interface was shown to contribute more to solar steam generation than to bulk fluid heating. Furthermore, the model of photothermal heating of plasmonic nanoparticle was established and the numerical results demonstrated the photothermal conversion process of plasmonic NPs from the light absorption to the heat dissipation into the bulk fluid.

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1. Introduction

Due to the increasing energy demand, solar energy has been selected as a promising green inexhaustible energy source [\(Lewis,](#page--1-0) [2007; Weinstein et al., 2015\)](#page--1-0). Traditional solar harvesting technologies are photovoltaics ([Atwater and Polman, 2010](#page--1-0)) and concentrated solar power (CSP) [\(Weinstein et al., 2015\)](#page--1-0), which are applied to generate electricity. Recently, more attentions have been focused on developing novel approaches for solar energy utilization such as seawater desalination ([Elimelech and Phillip, 2011;](#page--1-0) [Karagiannis and Soldatos, 2008\)](#page--1-0), photochemical reactions ([Chen](#page--1-0) [et al., 2016; Lewis, 2001; Naldoni et al., 2016](#page--1-0)), solar sterilization ([Neumann et al., 2013a](#page--1-0)), etc. Current methods of solar steam generation mainly rely on solar energy collection by black surface or cavity absorbers, with the thermal energy subsequently transferred to the working fluid either directly or via a high heat capacity intermediate carrier inside the absorbing tube to heat the bulk fluid to its boiling temperature ([Kundu and Lee, 2012; Lenert and](#page--1-0) [Wang, 2012; Weinstein et al., 2015\)](#page--1-0). This kind of indirect solar steam generation usually requires highly concentrated solar energy. Moreover, it suffers from high optical loss and thermal irradiation to the environment and high heat loss during convective heat transfer. To eliminate the energy loss due to indirect absorption, direct and volumetric absorption of solar energy by the working fluid can minimize the convection heat transfer losses and achieve higher efficiency [\(Ni et al., 2015; Otanicar et al., 2010](#page--1-0)).

Nanofluids, a kind of functional nanoparticle dispersions, have been widely studied since their discovery in 1990s ([Leong et al.,](#page--1-0) [2016](#page--1-0)). Recently, a number of experimental and theoretical investigations have found that nanofluid-based direct absorbing solar collectors (DASCs) exhibit excellent performance due to their photothermal characteristics ([Colangelo et al., 2015, 2013;](#page--1-0) [Karami et al., 2016, 2014; Liu et al., 2015; Luo et al., 2014;](#page--1-0) [Otanicar et al., 2010; Tyagi et al., 2009](#page--1-0)). Moreover, [Chieruzzi](#page--1-0) [et al. \(2017, 2015, 2016\)](#page--1-0) experimentally investigated and reviewed the development of nanofluids for thermal energy storage in solar energy applications and discussed the enhancement of fundamental thermal properties of nanofluids. [Shin and Banerjee \(2011\)](#page--1-0) reported an anomalous enhancement of specific heat capacity of

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nanofluids which was composited chloride salt eutectics and silica nanoparticles for thermal energy storage applications. [Otanicar](#page--1-0) [et al. \(2010\)](#page--1-0) studied direct absorption collectors with nanofluids containing carbon nanotubes, graphite, and silver nanoparticles, achieving an efficiency enhancement of up to 5%. [Taylor et al.](#page--1-0) [\(2011\)](#page--1-0) demonstrated that nanoparticles could absorb over 95% of incoming light by investigating the optical properties of different kinds of nanofluids. [Vakili et al. \(2016a, 2016b\)](#page--1-0) utilized graphite and graphene nanoplatelet nanofluid for direct absorption solar collectors exhibiting good photothermal properties at low temperature. [Colangelo et al. \(2015, 2013\)](#page--1-0) proposed a modified flat panel solar collector, and the results revealed that Al_2O_3 -distillated water could increase the thermal efficiency by up to 11.7%, as compared to pure water. [Gupta et al. \(2015\)](#page--1-0) experimentally investigated the volumetric absorption of thin film of $Al_2O_3-H_2O$ nanofluid and obtained high efficient enhancement of 22.1%.

In recent decades, plasmonic nanoparticles (metal materials or doped metal oxides) have attracted much interest and have been widely used in applications such as thermal cancer treatment ([O'Neal et al., 2004](#page--1-0)), magnetic recording [\(Challener et al., 2009\)](#page--1-0), and catalysis [\(Linic et al., 2011; Warren and Thimsen, 2012\)](#page--1-0) due to their thermal and thermo-optical properties caused by collective coherent excitation of the free electrons, which is also known as localized surface plasmon resonance (LSPR). Localized heat is generated by coupling the plasmonic nanoparticles and photons and is subsequently transferred to the surrounding medium. [Zhang et al.](#page--1-0) [\(2014\)](#page--1-0) and [Chen et al. \(2015\)](#page--1-0) investigated the photothermal conversion ability of gold and silver nanoparticle dispersions, respectively, obtaining high photothermal efficiency and bulk liquid temperature in both cases. [Du and Tang \(2016, 2015\)](#page--1-0) numerically investigated the optical properties of plasmonic nanofluids containing different shapes of Au NPs or agglomerated Au clusters and concluded that a blended nanofluids could enhance the solar energy harvesting.

Rather than heating the bulk fluid, plasmonic NPs can also generate nanovapor under continuous laser illumination [\(Baffou et al.,](#page--1-0) [2014\)](#page--1-0) or highly concentrated solar illumination ([Neumann et al.,](#page--1-0) [2013b](#page--1-0)). [Lombard et al. \(2015\) and Neumann et al. \(2013b\)](#page--1-0) demonstrated solar vapor generation using an Au-based dispersion, and achieved a solar steam device efficiency of 24% under the concentrated solar energy of 1000 sun (1 sun = 1 kW m $^{-2}$). [Ni et al. \(2015\)](#page--1-0) reported vapor generation efficiencies of up to 69% at 10 sun using a graphitized carbon black nanofluid and numerically demonstrated that the nanofluid steam generation phenomenon results from the global heating of bulk fluid. [Jin et al. \(2016\)](#page--1-0) performed steam generation experiments under sunlight of 220 Suns and revealed that the steam produced due to the highly non-uniform energy distribution in the system. [Wang et al. \(2016\)](#page--1-0) experimentally obtained a high evaporation efficiency through direct solar steam generation with carbon-nanotube nanofluid.

There are still few reports about the direct solar steam generation with plasmonic nanofluids, and a lack of systematic study on the effect of solar power intensities and plasmonic NP concentrations on solar steam generation performance. In this work, solar steam generation experiments were conducted with plasmonic nanofluids containing Au NPs under simulated solar light. The effect of Au NP concentration (5–178 ppm) and solar power intensity (1–10 sun) on evaporation performance were investigated by measuring the water weight loss and system temperature change. Furthermore, the optical properties of these nanofluids were studied by spectrometry, Mie theory, and finite difference time domain (FDTD) simulation. Finally, a model of heat generation through the light absorption and the heat dissipation to the surrounding medium was established and applied to demonstrate the mechanism of plasmon heating and localized heating during photothermal heating enabled by plasmonic NPs.

2. Experimental section

2.1. Materials

Tetrachloroauric acid (HAuCl₄; \sim 49–50% Au basis) and sodium citrate dihydrate (HOC(COONa)(CH₂COONa)₂.2H₂O, 99%, AR) were purchased from Aladdin Industrial Inc., Shanghai, China and were used without further purification. Doubly deionized water (DDI water) was produced by a Sartorius water purification system (arium \textdegree mini; 18.2 M Ω ; Göttingen, Germany).

2.2. Synthesis of Au NPs

Au NPs were synthesized by reduction of tetrachloroauric acid with citrate (\overline{I} i et al., 2007). HAuCl₄ (180.0 mg) was dissolved in DDI water (950 mL), and the solution was vigorously stirred at 500 rpm and heated for 1 h to reach its boiling point. Sodium citrate dihydrate (510.0 mg) was dissolved in DDI water (50 mL) and kept for further use. After boiling (100 °C) the HAuCl₄ solution for 20 min, the prepared citrate solution was rapidly added, inducing a color change from pale yellow to colorless and finally to wine red. Boiling was continued for another 20 min until the solution color was stable. The solution was cooled to room temperature with continuous stirring at 500 rpm.

2.3. Preparation of plasmonic nanofluids

To dispose of unreacted chemicals and concentrate the Au NP dispersion, centrifugation of the reaction mixture was performed at room temperature. The as-prepared aqueous Au NP dispersions were transferred into several 50-mL centrifuge tubes and centrifuged at 10,000 rpm for 1 h using Sigma laboratory centrifuges (2-16P, Sigma, Germany). The supernatant was subsequently removed, and the Au NP sediment was dispersed in 20 mL of DDI water. Finally, the concentrated dispersion was diluted to different concentrations for the solar steam generation experiment.

2.4. Solar steam generation experiment

The schematic of the localized heating of Au NPs dispersion for direct solar steam generation is shown in [Fig. 1a](#page--1-0). As Au NPs dispersion illuminated with simulated solar light on the top side of the fluid, multiparticle optical interactions in Au NPs dispersion were happened by the absorbing and scattering of incident photons. Eventually, the solar light was converted into thermal energy for the direct steam generation at the fluid-air interface. A schematic illustration of the solar steam generation setup is shown in [Fig. 1](#page--1-0)b. The setup features three main parts: simulated solar light generator, weight change monitor, and temperature increase monitor. As shown in [Fig. 1](#page--1-0)b, an acrylic tube with an inner diameter of 33 mm, inner height of 60 mm, and thickness of 3 mm was used as a testing chamber. In addition, five T-type thermocouples were inserted into the center of the tube to measure the temperature increase of the working fluid at different heights (start from the bottom of the tube: 10, 20, 30, 40, and 50 mm) due to light illumination. The testing chamber filled with working fluid (\sim 51.3 g) was placed on an electric balance (Practum313–1CN, Sartorius, Göttingen, Germany) that recorded its weight and exported the data to a computer every 1 min. A simulated solar light generator (CEL HXF300, CEAULIGHT, Beijing, China) illuminated the top of the testing chamber and the steam was generated due to the absorption of the solar light (at AM = 1.5). Meanwhile, the thermocouples (TT–T–40–SLE, Omega, US, accuracy of ± 0.5 °C) immersed in the working fluid were connected to the data acquisition system

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