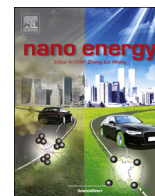




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Steam generation in a nanoparticle-based solar receiver

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ABSTRACT

Steam production is essential for a wide range of applications, and currently there is still strong debate if steam could be generated on top of heated nanoparticles in a solar receiver. We performed steam generation experiments for different concentrations of gold nanoparticles dispersions in a cylindrical receiver under focused natural sunlight of 220 Suns. Combined with mathematical modelling, it is found that the initial stage of steam generation is mainly caused by localized boiling and vaporization in the superheated region due to highly non-uniform temperature and radiation energy distribution, albeit the bulk fluid is still subcooled. Such a phenomenon can be well explained by the classical heat transfer theory, and the hypothesized 'nanobubble', i.e., steam produced around the heated nanoparticles, is unlikely to occur under normal solar concentrations. For future solar receiver design, attention should be paid to focus and trap more solar energy at the superheated region while minimizing the temperature rise of the bulk fluid.

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1. Introduction

Steam production is essential for a wide range of applications from large scale electricity generation, energy storage, desalination systems and refrigeration units to compact small scale systems such as sterilization and clearing [1–4]. Conventionally steam is produced by the combustion of fossil fuels or direct heating from electricity, which is environmentally unfriendly. Employing solar energy, an abundant, clean and renewable energy source, for steam production is a rapidly developing area [5–8]. Currently solar-based steam production (i.e., either solar trough or solar tower systems) is based on heating a bulk fluid to its boiling temperature under high optical concentrations. The steam generation efficiency heavily relies on the surface temperature and radiation properties of the absorber, whose high temperature needed for bulk steam production leads to large heat loss to the ambient and low energy efficiency.

It has been reported recently that certain nanoparticles, especially those with Localized Surface Plasmon Resonance (LSPR) properties [9,10], can absorb solar energy efficiently in a liquid medium [11–14]. For an aqueous nanoparticle dispersion [15], it would lead to a rapid increase of the particle temperature and steam production, albeit the bulk fluid was still in the subcooled

state [16,17]. For example, the research team from Rice University showed that by using a very dilute gold nanoparticles dispersion, i.e., 16.7 ppm, under focused sunlight via a Fresnel Lens, rapid steam production was realized while the bulk fluid temperature was still at $\sim 6^\circ\text{C}$ [11]. The calculation showed that the steam generation efficiency was reached up to 80%, and only a small portion of the solar radiation was used to increase the bulk fluid temperature. Similar to the concept of energy localization on the surface [18], it appears that solar energy was localized by the nanoparticles. It was further hypothesized that rapid heating of nanoparticles produced nanobubbles immediately around the nanoparticles, and the rise of nanobubbles to the top surface of the liquid realized the release of the vapor produced [19–21]. Subsequent simulation work [11,16,17,22] showed the possibility of nanobubble formation based on a non-equilibrium phase change assumption.

The heating of nanoparticles and formation of nanobubbles have become an intensive research topic in the medical area. It has been confirmed both experimentally and theoretically [13,14,16,23–25] that under an intensive laser heating (i.e. $> 1000\text{ MW/m}^2$), bubbles can be generated around the heated nanoparticles [26,27]. By controlling the laser power and pulse appropriately, the growth and contraction of bubbles can be very fast, which is associated with the propagation of pressure waves that could bring thermal-mechanical damage to surrounding cells at a dimension much larger than that of a single nanoparticle [28]. However, it is still unclear if bubbles can be formed under a relatively low heat flux provided by concentrated sunlight (i.e.,

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typically $< 1 \text{ MW/m}^2$), and a few concerns have been raised recently. For instance, Ni et al. [29] showed that there were no nanobubbles produced under a solar concentration of 10 Suns, and suggested that the classical global heating may be responsible for the steam generation. Through a molecular dynamics simulation study, Chen et al. [19] also showed that it was difficult to form nanobubbles under continuous heating conditions even under a high heating power.

It shall also be of note that most of the solar steam generation experiments employed only one point temperature measurement [11,29,30], without knowing the temperature distribution of the fluid, which may lead to misleading or unconvincing conclusions. Clearly there is still a critical lack of both strict experimental evidence and well-accepted mechanism analysis in the solar steam generation. Aiming to address these contradictions, steam generation using gold nanoparticle dispersions with different concentrations in a cylindrical tube under focused natural sunlight was investigated experimentally, and a 3-D mathematical model was also established to reveal the non-uniform temperature distribution inside the nanoparticle dispersions. We revealed that steam generation during the heating up stage was mainly caused by localized boiling and vaporization in superheated regimes due to a highly non-uniform temperature distribution, albeit the bulk fluid is still subcooled. Such a phenomenon can be explained by the classical heat transfer theory and the hypothesized nanobubble, i.e., steam produced around heated particles, was unlikely to occur under normal solar concentrations.

2. Experimental details

2.1. Nanoparticles fabrication and characterization

A one-step method [31] was employed to produce stable gold nanoparticles (GNPs) dispersions. First, $5 \times 10^{-6} \text{ mol HAuCl}_4$ (Sigma-Aldrich) was dispersed into 190 ml DI water in a three-necked flask, then a magnetic blender with a heating source was used to stir the liquid until the occurrence of boiling. 10 mins later, 10 ml aqueous sodium citrate (Sigma-Aldrich) solution with a mass concentration of 0.5% was added into the prepared HAuCl_4 solution. The mixed solution turned dark blue within 30 s, and the final color became wine red after being heated for an additional 20 mins. The GNPs dispersions maintained good stability for over two months, and were used for the experiments without further purification and separation. Gold nanoparticles' size and shape were characterized (Fig. 1A and B) by the Transmission Electron Microscopy (TEM) (FEI Tecnai TF20: FEGTEM Field emission gun TEM/STEM fitted with HAADF detector, Oxford Instruments INCA 350 EDX system/80 mm X-Max SDD detector and Gatan Orius SC600A CCD camera). A dynamic light scattering (DLS) device (Malvern nanosizer) was employed to identify the particle size distribution, which is presented in the [Supporting Information](#).

2.2. Experimental setup

The experiments were performed under focused natural sunlight (Fig. 1C and D). Diluted GNPs dispersions (with concentrations of 1.02 ppm, 5.1 ppm and 12.75 ppm) and DI water were placed into four cylindrical tubes (i.e., inner diameter of 25 mm and length of 300 mm), respectively. The tubes were custom-made from high temperature resistant quartz, and vacuum interlayers were employed to reduce the convection heat loss to the ambient as much as possible (Fig. 2). The outer tube with a diameter of 60 mm had two small-bore pipes, which were used to fix the thermocouples. Fresnel lenses (400mm \times 400mm) with a 620 mm focal distance were used to focus the natural sunlight. The smallest

focused spot has a diameter of 30 mm, and the focused solar intensity in the experiments was 220 Suns. A solar radiation intensity sensor (SPN1, Delta-T Devices) with a measurement uncertainty of 2% was employed to measure the solar intensity.

In order to investigate non-uniform temperature distribution within the fluid, three type T thermocouples (Omega TT-T-40-SLE) with precision of $\pm 0.5^\circ\text{C}$ were placed in the bottom, middle and top of the test sample fluids, respectively. Another two thermocouples were placed inside and outside the cylindrical tube to measure the steam and ambient temperatures, respectively (Fig. 2). A microbalance (OHAUS Adventurer) was employed to measure the mass change of fluid when illuminated, where a water cooling system was used to condense the generated steam (Fig. 1D). Before the experiments, all the four cylindrical tubes were cleaned carefully with pure water at ambient temperature. These tubes with test sample fluids were then heated simultaneously under the same solar conditions (220 Suns). Due to the movement of the Sun and the change of liquid position because of steam generation, the focus point was manually adjusted to keep it on the fluid.

3. Results and discussions

3.1. Temperature profile

Once the tube was illuminated under 220 Suns, fluid temperatures rose immediately (Fig. 3), and main observations can be summarized as follows:

- (1) It was difficult for DI water to reach boiling under such a solar intensity (i.e. 220 Suns). For DI water (Fig. 3D), the highest temperature was increased to only 65°C (TC3) after 5 min' illumination. One position reached 90°C after 10 min' illumination, then it remained nearly constant, indicating the attainment of a steady state where the heat loss was equal to the absorbed solar radiation energy.
- (2) All GNPs dispersions reached the boiling temperature fast and then remained unchanged at that value. Increasing the volume concentration could reduce the time required to reach the boiling point. For 1.02 ppm GNPs dispersion (Fig. 3A), it took more than 3 min for all the three measured positions to reach the boiling point. While for higher concentrations such as 12.75 ppm (Fig. 3C), it was reduced to only 90 s.
- (3) Large temperature differences existed within the fluid before reaching the boiling point, and the temperatures at the measured positions was highly non-uniform for all the sample fluids. For instance, an impressive temperature difference was observed, i.e., 46.5°C between TC1 and TC3, in less than 1 min's illumination for 12.75 ppm GNPs dispersion. However, for all GNPs dispersions, the temperature non-uniformity became much smaller after reaching the boiling point.
- (4) Steam can be generated under subcooled conditions and was highly particle concentration dependent. For 1.02 ppm and 5.1 ppm GNPs dispersions, appreciable steam temperature increase was only detected when the bulk fluid temperature reached approximately the boiling point (as shown by arrows in Fig. 3). However, for 12.75 ppm GNPs dispersion, almost immediately air temperature rise inside the tube was observed, indicating that vapor was generated rapidly. At that time, all the three thermocouples indicated that the bulk fluid temperature was still very low and impossible for boiling to happen. This suggested that vapor was produced when the bulk fluid was in the subcooled state, similar to the results reported by the research group from Rice University [11].

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