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A comparative study of direct absorption nanofluids for solar thermal applications

Aimen Zeiny[a, c], Haichuan Jin[b], Lizhan Bai[b], Dongsheng Wen[b, a *]

[a] School of Chemical and Process Engineering, University of Leeds, Leeds, LS2 9JT, UK

[b] Laboratory of Fundamental Science on Ergonomics and Environmental Control, School of Aeronautic Science and Engineering, Beihang University, Beijing 100191, PR China

[c] Department of Mechanical Engineering, University of Kufa, Najaf, Iraq

Tel: +44 113 3431299

[*] Corresponding author and E-mail: D.Wen@leeds.ac.uk

Abstract

Direct absorption nanofluid has been introduced to as an effective alternative to increase the solar thermal conversion efficiency. Hybrid nanofluids were also recently proposed to broaden the absorption spectrum, however, a comparative assessment of the performance of commonly used nanomaterials for solar energy harness is still lacking. This study performed a well-controlled experiment for three different categorised particles, i.e., gold, copper, carbon black nanofluids and their hybrids, and assessed their performance in terms of photothermal conversion efficiency (PTE), specific absorption rate (SAR) and materials cost. Contrary to previously reported, the PTE was not increased by blending different nanofluids with different absorbance peaks, mainly due to the dilution of nanoparticle concentration. Though having high SAR, the high cost of gold prevents its practical use, and carbon black appears to be more feasible. The theoretical PTE can be well predicted by the optical properties of the nanofluids used.

Keywords: Direct absorption, nanofluid, nanoparticle, solar energy, solar thermal system.

1. Introduction

Solar energy has been claimed as the energy of our future but comes with many challenges to overcome such as the cost and the efficiency (Otanicar et al., 2010, Gupta et al., 2015). Solar thermal system, which typically has absorbing plate with fluid running inside pipes, is a common way of utilising solar energy. Its efficiency is limited by not only how efficiently the solar energy is captured by the absorbing plate, but also how effectively the absorbed energy is transferred to the running fluid (Otanicar et al., 2010). This surface-limited thermal energy transfer process limits the solar energy utilisation efficiency especially for high-temperature applications such as solar thermal power plants (Lenert and Wang, 2012).

Direct absorption solar collector (DASC) is proposed in the 1970s. In this approach, the solar energy is directly absorbed by the working fluid by seeding certain particles in it (Minardi and Chuang, 1975, Arai et al., 1984, Bertocchi et al., 2004). Early studies were focused on micrometer-sized particles, and using nanoparticles is a recent development (Zhang et al., 2015, Zhang et al., 2014, Otanicar et al., 2010, He et al., 2013). Many studies have been conducted and most of these studies were based on the optical properties and characterized by the extinction coefficient (Mercatelli et al., 2011, Otanicar et al., 2009, Otanicar et al., 2010, Sani et al., 2010, Sani et al., 2011, Taylor et al., 2011). The extinction coefficient is a key factor, which determines the property of a nanofluid of light absorbing and scattering but may not represent the actual photothermal conversion process (Zhang et al., 2014).

The photothermal conversion efficiency of different nanoparticles, ranging from metal (such as Au, Ag, Cu, and Al), metal oxides (such as CuO, TiO_2, Al_2O_3, and Fe_2O_3) and carbon (such as Graphite, carbon nanotubes (CNTs), and carbon nanohorns (CNHs)), have been investigated under the laboratory and outdoor conditions. Plasmonic nanoparticles (such as Au and Ag) have attracted intense interest due to the effect of surface plasmon resonance (SPR), which usually occur in the visible light spectrum, which represents almost 40% of the total solar energy (Gueymard, 2004) but weakly absorbed by most of the heat transfer fluids. Within these studies, Zhang et al. (Zhang et al., 2014) showed that using 6 ppm (~116 mg/l) aqueous gold nanofluid could enhance the photothermal conversion efficiency by ~80% and reached a specific absorption rate (SAR) of ~1kW/g under a solar simulator. An enhancement of 144% in the photothermal conversion efficiency and ~0.6 kW/g were achieved by using 6.5ppm (~68 mg/l) silver aqueous nanofluid in the outdoor conditions

(Bandarra Filho et al., 2014). However, the enhancement of absorption of plasmonic nanofluids has unavoidably a narrow bandwidth owing to their resonance characteristic (Jeon et al., 2016).

To achieve a broadband absorption, blending of different absorption peak nanofluids was suggested by some researchers. A blend of spherical gold nanoparticles with two different size gold nanoshells was proposed by Cole and Halas (Cole and Halas, 2006). They determined, theoretically, the ideal fractions of the blended nanofluids that match the AM 1.5 solar spectrum, and revealed that mixing 35.9% gold nanospheres of 32 nm radius, 22.8% gold nanoshells of 28 and 42 nm core and shell radii, and 41.3% gold nanoshells of 47 and 58 nm core and shell radii respectively, can achieve a photothermal conversion efficiency of ~84%. Lee et al. (Lee et al., 2012) investigated theoretically the efficiency of a direct solar collector using a hybrid core-shell gold nanofluid generated by mixing four different core and shell sizes. An efficiency of 70% was achieved by 0.05 vol.% (~ 3220 mg of gold/l) hybrid nanofluid. However, although the tunability of the optical properties of such core-shell nanoparticles is achievable, the difficulty in manufacturing this type of nanoparticle makes it not suitable for further solar applications (Jeon et al., 2016). Very recently, a blend of three different aspect ratios of gold nanorods was suggested by Jeon et al. (Jeon et al., 2016) to optimise the solar absorption efficiency. The photothermal performance of the blended nanofluids was checked by measuring the temperature rise with time. However a common problem among these studies is the hybrid nanofluids had much higher concentration than their individual counterparts due to the mixing. A fair comparison of the photothermal performance under a given concentration between the hybrid and its original nanofluids is still lacking . Such a comparison is crucial to check the feasibility of hybrid nanofluids.

In most of the previous studies, a uniform temperature distribution within a nanofluid was assumed although the effect of the optical path was not negligible, and one temperature was used to calculate the photothermal conversion efficiency (He et al., 2013, Bandarra Filho et al., 2014, Chen et al., 2015, Yousefi et al., 2012). However, the non-linearly reduction in the radiative intensity along the depth of the nanofluid should cause a temperature difference within the nanofluid (Jin et al., 2016c, Jin et al., 2016a). Neglecting this temperature difference will lead to an inaccurate calculation of the efficiency (Jin et al., 2016a). Moreover, most of the published work was based on only one particular type of particles, and a comparative assessment of the performance of commonly used nanomaterials for solar energy harness is still lacking. The effect of these nanomaterials needs to be investigated at the same concentration and under similar operating conditions to reach a fair comparison. In addition to the efficiency, the cost has to be considered very carefully for any

practical application. For the purpose of comparison, some estimation of the cost of a unit thermal power generation ($/kW) from different nanoparticles is preferred.

This study aims to overcome the issues reviewed above and perform a well-controlled experiment for three different categorised particles, i.e., gold, copper, carbon black nanofluids and their hybrids. A Class AAA solar simulator was used to ensure stable and uniform radiation intensity, and samples of ~3 mm thick were used to eliminate the effect of the absorbing path length. Photothermal conversation efficiencies (PTE), specific absorption rates (SAR) and cost of a unit thermal power generation ($/kW) were calculated from the recorded temperature rise of different nanofluids. In addition, estimated solar photothermal conversion efficiencies (i.e., without the need to measure the temperature rise of the nanofluids) were calculated from the spectral extinction coefficients measured by a spectrophotometer.

2. Experimental approach

2.1 Gold nanofluids preparation

In this study, gold nanofluid was synthesised by the citrate reduction method as reported by Chen and Wen (Chen and Wen, 2011) and Zhang et. al (Zhang et al., 2014). Typically, 100 ml of 5 mM $HAuCl_4$ solution was mixed with 100 ml of 10 mM trisodium citrate solution. Then, the resultant mixture was heated to the boiling temperature until its colour became wine red. After that, the resultant was put into a sonication bath at 80^{o}C for 3 hours. The synthesised gold nanoparticles were left for 24 hours at the room temperature and then purified by the membrane dialysis method. In this process, the gold nanofluid was put in a membrane tube with a pore size of 2-3 nm in diameter to allow a smooth diffusion of ions and keep the gold nanoparticles inside the tube. The membrane tube was located in a beaker filled with DI water of 2000 ml and stirred by a magnetic stirrer. The DI water was changed twice a day for ten days. The gold nanofluid is shown in Figure 1.

2.2 Copper and carbon black nanofluids preparation

Copper and carbon black nanofluid were prepared by the two-step method, i.e. by dispersing a certain amount of pre-synthesised nanopowder to a hosting liquid, i.e., DI water in this work. The copper nanopowder was purchased from Sigma-Aldrich Corporation, and carbon black nanopowder was purchased from Alfa Aesar. Dispersing agents of trisodium citrate (TSC) aqueous solution 0.5 M and Gum- Arabic (GA) powder were added to DI water at 2 vol. % and 0.5 wt. % respectively to

prepare the hosting liquid for copper nanofluids, and dispersing agent of Tween was added to DI water at 0.04 vol. % for carbon black nanofluid. The hosting liquid magnetically stirred when controlled amount of nanopowder was added. After 15 minutes, the sample was put into an ultrasonication bath. For copper nanofluids, the ultrasonication time was one hour, and for carbon black nanofluids, an ultrasonication bath for 30 minutes was used, followed by a powerful probe sonicator for extra 30 minutes. The copper and carbon black nanofluids are shown in Figure 1.

A flame atomic absorption spectrometer (AAS) was used to measure the concentration of the prepared nanofluids. For gold nanofluid, the original concentration was determined as 250 mg/l. The difference between AAS measured and pre-determined concentration of copper nanofluid was 3.76%, which shows the reliability of the method. Different dilutions were prepared from the stock nanofluids. The particle size distribution and zeta potential of the different nanofluids were measured by a zetasizer (Malvern) based on dynamic light scattering (DLS), as shown in Figure 2. A UV-Vis Spectrophotometer (UV-1800, SHIMADZU) was used to measure the absorption capability of the different nanofluids. The results are presented in Figure 3, which reveal an excellent agreement between the absorbance results and Beer's law, indicating a linear relationship between absorbance and solution concentration. It is clear that gold nanofluids have good absorbance in the range 300-600 nm wavelength and the peak value is around 528 nm, which is due to the local surface plasmon resonance, while the peak value of the absorbance of the copper nanofluids is around 740 nm. However, the absorption capability of the carbon black nanofluids is better than the gold and copper nanofluids over the whole spectrum.

2.3 Photothermal conversion experiment

The photothermal conversion experimental setup is shown in Figure 4. A sun simulator (ORIEL® Sol3A™ CLASS AAA SOLAR SIMULATOR) was used as a light source to minimise the uncertainties under direct sunlight. This sun simulator provides a radiation spectrum matches the solar spectra, and the intensity can be varied by using suitable filters. This device is certified to IEC 60904-9 2007 edition, JIS C 8912 and ASTM E 927-05 standards. The performance parameters of the sun simulator are; spectral match between 0.75 – 1.25%, non-uniformity < 2% and temporal instability < 0.5%. By using a precise micro-pipette, a 3 ml sample was put in a Petri dish (35 mm diameter) to get on a thin layer of the nanofluid (~3 mm) to minimise the temperature gradient within the sample. The Petri dish was located on the bottom of an upside down beaker in the centre spot of the solar simulator, and it was covered by a thin glass to minimise the losses from the sample. A K-type thermocouple (Omega 5TC-TT-K-36-36, a precision of ±0.5 ^{o}C) was used to measure the

temperature of the centre of the sample. The transient bulk temperature data was recorded to a PC via data acquisition hardware under the LabVIEW environment. Preliminary tests with five thermocouples located at different positions on the bottom of the Petri dish showed that the space variation of the sample temperature was negligible. Furthermore, by using two thermocouples, one is just under beneath the sample level and the other at the bottom of the sample, the highest temperature difference was less than 0.5 °C for the highest concentration sample. Therefore, using one thermocouple located at the centre of the sample is very reliable.

3. Results and Discussion

The nanofluid samples were subjected to 1000 W/m^2 radiation from the solar simulator for 300 s during which the temperature was being recorded. The photothermal conversion efficiency is defined as the ratio between the energy stored in the nanofluid and the total incident radiation:

$$\boldsymbol{PTE} = \frac{(\boldsymbol{c_w m_w \Delta T_w + c_{np} m_{np} \Delta T_{np}})/\Delta \boldsymbol{t}}{\boldsymbol{IA\Delta T}} \quad (1)$$

where c_w and c_{np} are the specific heat of water and the nanoparticles; m_w and m_{np} are the mass of water and the nanoparticles respectively; ΔT_w and ΔT_{np} are the water and nanoparticles temperature rise in the Δt time interval; I is the radiation intensity; and A is the illumination area of the nanofluid. The temperature difference between the nanoparticles and the hosting fluid (water) is negligible for low intensity continuous radiation (Keblinski et al., 2006, Zhang et al., 2014), and the nanoparticles mass is negligible comparing to the hosting fluid mass. Therefore, Eqn. 1 can be re-written as:

$$\boldsymbol{PTE} \approx \frac{\boldsymbol{c_w m_w}}{\boldsymbol{IA}} \frac{\boldsymbol{\Delta T_{nf}}}{\boldsymbol{\Delta t}} \quad (2)$$

where ΔT_{nf} is the temperature difference rise of the bulk nanofluid.

The transient temperature difference and photothermal conversion efficiency (PTE) curves are shown in Figure 5 for gold, copper and carbon black nanofluids respectively. It is evident that the temperature difference rise for each nanofluid increases as the time increases. Also, it is clear that ΔT_{nf} increases as the nanoparticle concentration increases. This is due to the enhancement of the water absorption of the radiation by the nanoparticles. Returning to Eqn. 2, the PTE is proportional to the ratio $(\frac{\Delta T_{nf}}{\Delta t})$, or in other words, PTE is a function to the rate change of the temperature. Thus the PTE decreases as the time increases, as shown in Figure 5.

Due to the negligible effect of the length of the light path within the sample (sample thickness is ~ 3mm), and according to the Beer's law, which indicates that absorbance of a sample is linearly

proportional to its concentration, it is expected that ΔT_{nf} is linearly proportional to the nanoparticles concentration. Indeed, that is well presented in Figure 6, and it is clear that the rate change of the temperature rise with respect to the concentration increases as the time increases. This is because more radiative energy is captured and converted to thermal energy with time. As a consequent, the PTE increases linearly as the concentration of the nanoparticles increases as shown in the inset of Figure 6.

Practically, the improving in the PTE of nanoparticles seeded working fluid comparing to the PTE of the hosting fluid ($enhancement = \frac{PTE_{nf} - PTE_w}{PTE_w}.100\%$) is the fruit. A comparison between the gold and copper nanofluids is presented in Figure 7. Obviously, the higher the nanoparticles concentration, the higher the photothermal conversion enhancement is. The maximum enhancement for gold nanofluid is ~72% at 150 mg/l concentration, and for copper nanofluid is ~ 100% at 3000 mg/l. However, it seems that the PTE of 60 mg/l gold nanofluid approximately equal to 750 mg/l copper nanofluid, 90 mg/l gold nanofluid approximately equal to 1250 mg/l copper nanofluid, and 150 mg/l gold nanofluid is equal to 2000 mg/l copper nanofluid.

Blending different plasmonic nanofluids with different absorption peaks was suggested by Cole and Halas (Cole and Halas, 2006), Lee et al. (Lee et al., 2012), and Jeon et al. (Jeon et al., 2016) to improve the radiative absorption with broader absorption peaks. However, none of them compared the PTE of the hybrid nanofluid with that of the original nanofluids under the same concentration. In this work, hybrids from blending gold (absorption peak at ~528 nm wavelength) and copper (absorption peak at ~740 nm wavelength) were prepared. Firstly, 1 ml gold nanofluid was blended with 1 ml copper nanofluid, and then an absorbance spectrum test was performed for the hybrid. The results are shown in Figure 8. Indeed, the width of the absorbance peak became broader for the hybrid, but the absorbance value apparently became lower. It is logical because mixing different nanofluids together leads to the dilution of the original nanofluids due to the increase in the overall volume of the hybrid. For instance, in a hybrid of 1 ml gold nanofluid with 1 ml of copper nanofluid, the concentration of gold nanoparticles becomes half of the original, and so as to the copper nanofluid (i.e., this is true if there is no reaction and/or aggregation between the blended nanofluids). Good agreement is found between the absorbance spectrum of the hybrid measured by the spectrophotometer (experimental) and the average absorbance of the original nanofluids (theoretical). Such results were quite different to what reported earlier (Lee et al., 2012, Cole and Halas, 2006). Though a high solar collector efficiency was reported by using a hybrid in Jeon's

experiment (~71%) (Jeon et al., 2016), the result is misleading due to a nearly doubled hybrid particle concentration used.

To check this thermally, photothermal conversion tests were conducted for three hybrids. These hybrids were prepared by mixing equal portions of gold and copper nanofluids that showed approximately the same PTE enhancement, i.e., 60 mg/l gold nanofluid with 750 mg/l copper nanofluid, 90 mg/l gold nanofluid with 1250 mg/l copper nanofluid, and 150 mg/l gold nanofluid with 2000 mg/l copper nanofluid respectively. The results are shown in Figure 10, and it is clear that the temperature rise curves are almost identical for the hybrids and their original ingredients. Using hybrid nanofluids does not produce any additional benefits. Comparing to other materials, carbon-based nanofluids have the most desired absorption spectrum (see Figure 3 (C)). With its super absorbance at low concentrations (less than 100 mg/l), carbon black nanofluids appear the most feasible choice for solar energy harvesting, where an enhancement of ~125% was achieved by 100 mg/l carbon black nanofluid, as shown in Figure 10.

From the previous results, it is clear that for the same nanoparticles concentration, the temperature rise of gold nanofluids is higher than that of copper nanofluids. Therefore, to quantify the ability of a certain nanoparticle to generate thermal power, the specific absorption rate (SAR) is used, which indicates the amount of thermal power generated by a unit mass of nanoparticles (Bandarra Filho et al., 2014, Zhang et al., 2014):

$$\boldsymbol{SAR} = \frac{[(c_w m_w + c_n m_n)\Delta T_{nf} - c_w m_w \Delta T_w]/\Delta t}{m_n} \tag{3}$$

However, the cost of the nanofluids is crucial for solar applications, especially applications that need a large amount of working fluids. So, to investigate this factor, the cost of generating a unit thermal power is employed:

$$\frac{\$}{kW} = \frac{cost_n[^{\$}/_{g}]}{SAR[^{kW}/_{g}]} \tag{4}$$

where $cost_n$ is the cost of the nanoparticles used in synthesizing the nanofluids. The price of gold and copper was taken from the international markets for metal stocks, which represents the lowest cost to generate one kW thermal power, while the price of carbon black was taken from the supplier (Alfa Aesar). Figure 11 shows the SAR and $/kW of both gold, copper and carbon black nanofluids. It is obvious that gold nanofluids have higher SARs than copper nanofluids (ten folds), but the cost

of generating 1kW is much higher than that of copper nanofluids (i.e., the cost of using the lowest gold nanoparticles concentration, 30 mg/l, is higher than $100, while the cost of using the highest copper nanoparticles, 3000 mg/l, is less than $ 0.3). However, the SAR for the carbon black nanofluids is higher than that of gold nanofluids and, at the same time, $\frac{\$}{kW}$ is comparable with that of copper nanofluids.

The prediction of the maximum PTE can be done by relying on the results of the absorbance spectrum tests, where the ratio of the transmitted intensity to the incident intensity defines the transmittance (T):

$$\boldsymbol{T} = \frac{I_t}{I_i} \tag{5}$$

$$\boldsymbol{A} = -\boldsymbol{logT} \tag{6}$$

where A is the absorbance. The scattering effect can be neglected because $\alpha \ll 1$, where α is the particle size parameter and equals to $\frac{\pi D}{\lambda}$, D is the nanoparticle's diameter and λ the wavelength (Taylor et al., 2011, Lenert and Wang, 2012, Khullar et al., 2012), and therefore,

$$\boldsymbol{I_{abs} = I_i - I_t = I_i(1 - T)} \tag{7}$$

where I_{abs} is the absorbed intensity. The optical efficiency (maximum PTE) is the ratio of the absorbed intensity to the incident intensity:

$$\boldsymbol{PTE_{max}} = \frac{\int_0^\infty \boldsymbol{I_{\lambda,abs}\, d\lambda}}{\int_0^\infty \boldsymbol{I_{\lambda,i}\, d\lambda}} = \frac{\int_0^\infty \boldsymbol{I_{\lambda,i}(1 - e^{-Ay})\, d\lambda}}{\int_0^\infty \boldsymbol{I_{\lambda,i}\, d\lambda}} \tag{8}$$

where y is the light path length. The solar spectral irradiance, which represents air mass (AM) 1.5 was taken from the literature (Gueymard, 2004).

Due to the limitation of the spectrometer used in this study (320 nm $\leq \lambda \leq$ 1060 nm), and because water is almost transparent over this range (see Figure 3(A)), the calculated PTE_{max} was assumed to represent the effect of nanoparticles only. Therefore, the PTE enhancement can be re-written as:

$$\boldsymbol{enhancement_{max}} = \frac{(\boldsymbol{PTE_{max} + PTE_w}) - \boldsymbol{PTE_w}}{\boldsymbol{PTE_w}} . \boldsymbol{100}\% \tag{9}$$

A comparison between the maximum enhancement predicted from Eqn. 9 and the photothermal conversion enhancement is shown in Figure 12. The results show a linear behaviour as expected. It is clear that carbon black (CB) nanofluids have the highest enhancement comparing to gold and copper nanofluids, due to their broad and high absorbance spectrum.

4. Conclusion

In order to enhance the solar photothermal conversion performance based on the direct absorption concept, gold, copper, gold-copper hybrid, and carbon black nanofluids with different concentrations were prepared and characterised in this work. Extensive experiments were conducted with various nanofluids under a solar simulator, and important conclusions can be drawn as follows:

- Seeding water with nanoparticles enhances the solar photothermal conversion efficiency, and this enhancement is linearly proportional to the nanoparticles concentration, which obeys Beer's law.
- The solar energy absorption efficiency does not increase by blending different nanofluids with different absorbance peaks. Although the blending broadens the peak's width, it reduces the absorption peak value due to the dilution in particle concentration.
- Considering both photothermal conversion efficiency and potential economic cost, carbon black nanofluid is a suitable candidate, while gold is not.
- Using optical properties of a nanofluid could predict its maximum solar photothermal conversion efficiency.

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