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PHOTOTHERMAL ENERGY CONVERSION IN LIQUID NANOPARTICLE SUSPENSIONS

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ABSTRACT

Liquid nanoparticle suspensions, popularly termed "nanofluids," have been the subject of numerous investigations because of their interesting thermal transport properties. Their propensity to scatter and absorb electromagnetic radiation enables other applications that can take advantage of both their radiative and thermal transport properties. In particular, we are working to develop direct-absorption solar thermal collectors in which nanofluids serve to absorb incident sunlight, thus heating the fluid directly and more efficiently than conventional solar collectors. Our experimental results, in which we irradiate nanofluids with a continuous-wave laser, demonstrate that boiling can be induced at lower incident light fluxes compared to a thin layer of pure water in front of a black absorptive backing. These findings suggest that improved solar energy conversion systems can be developed, including solar-driven direct-steam generators.

INTRODUCTION

Liquid nanoparticle suspensions, popularly called "nanofluids" in the heat transfer literature, have been widely investigated for their interesting thermal conduction and convection properties (see, e.g., **[1] [2] [3] [4] [5] [6]**). The radiative properties of nanofluids, however, have been much less studied **[7]**, even though it is readily apparent to anyone who has observed a prepared nanofluid that the nanofluid strongly scatters and absorbs visible light. This observation generated the concept that nanofluids can be used as direct volumetric absorbers for solar radiation (**[8] [9] [10] [11] [12] [13]**), leading potentially to more efficient solar energy conversion which we term *photothermal energy conversion*. The basic idea is schematically presented in Fig. 1. A flowing nanofluid is exposed to incident light, which can be concentrated or not, through a transparent cover. Absorption of light within the nanofluid, largely by the nanoparticles, leads to a temperature rise in the nanofluid that can then be exploited

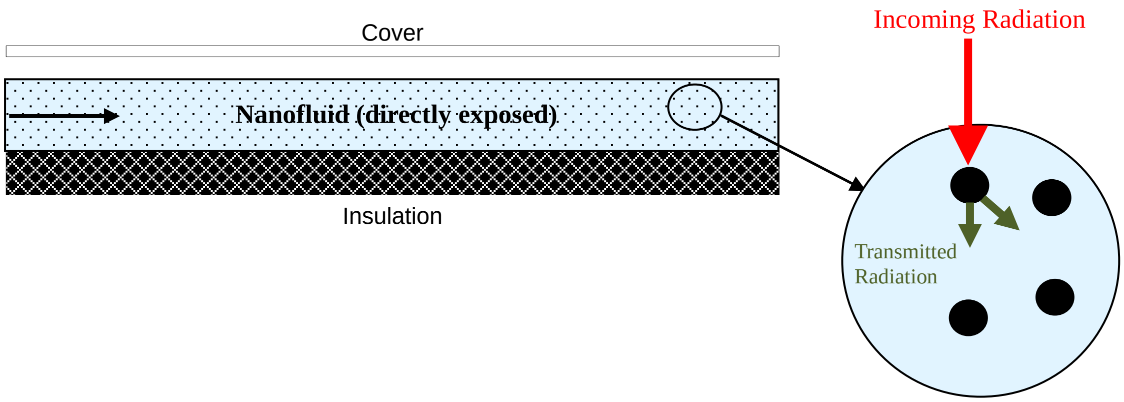

Fig. 1 **Schematic diagram of a nanofluid-based volumetric direct-absorption solar receiver.**

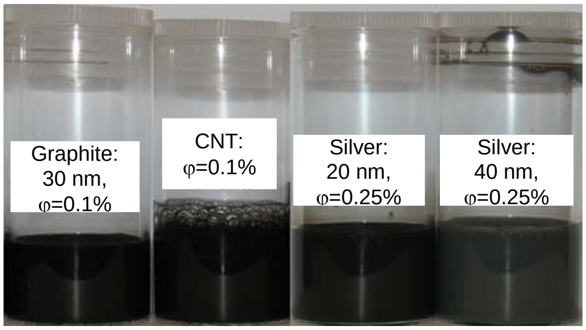

Fig. 2 Representative nanofluid samples tested in micro collector (CNT = carbon nanotubes)

for energy conversion. Our continuing research demonstrates that not only can sensible nanofluid temperature increases occur, but also boiling can take place around the nanoparticles. This gives rise to the possibility that direct-absorption nanofluids, like those under investigation here, can be utilized for concentrating solar direct-steam generation, a promising direction for concentrating solar power (CSP) technology (**[14] [15] [16] [17]**).

Here we review some of our recent results, with emphasis on boiling induced by laser heating of the nanofluid. A brief discussion of the radiative properties of nanofluids is also presented, and we conclude with suggestions for future directions.

NANOFLUIDS AND SOLAR THERMAL ENERGY

Photographs of representative nanofluids are presented in Fig. 2. From these pictures it is clear that even nanoparticle volume fractions, φ, as low as $\varphi = 0.1\%$, yield very dark, strongly scattering/absorbing nanofluids. Such fluids can then serve to absorb sunlight directly, as opposed to the conventional approach in which a solid receiver surface absorbs sunlight, and then has to transfer that energy to a fluid. For liquids, this can be accomplished by the introduction of a black dye, a concept dating back to the 1970's (see, e.g., [18]), or by creating a colloidal suspension (see, e.g., [19] [20] [21]). Alternatively, particles can be suspended in gases for high-temperature solar applications (see, e.g., [22] [23] [24] [25] [26]). Following these approaches, we suggest that nanofluids can be utilized for solar thermal energy conversion, as presented schematically in Fig. 3, which shows how concentrated sunlight is absorbed directly in the nanofluid, which can then serve as the heat source to drive a heat engine or other thermal process. Another intriguing possibility is direct steam generation in water-based nanofluids, which we discuss in the following section.

Does the use of direct-absorption nanofluids lead to higher solar collector efficiencies? The answer is yes:

- For flat-plate (nonconcentrating) collectors, calculated efficiencies were as much as 10% higher, on an absolute basis, than comparable conventional flat-plate collectors [8].

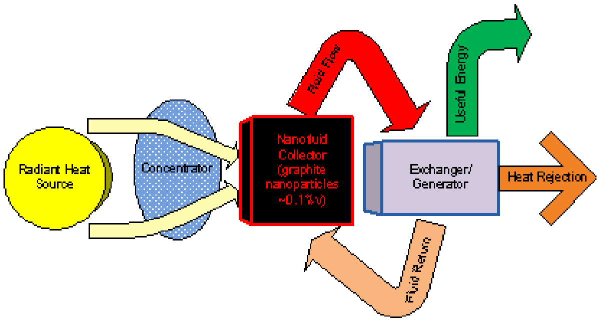

Fig. 3 Proposed nanofluid-based photothermal energy conversion

- Experimental data on micro-scale solar collectors yielded a maximum collector efficiency of 57% for a nanofluid consisting of $\varphi = 0.25\%$ 20-nm Ag nanoparticles in water, compared to a maximum collector efficiency of 52% for pure water flowing over a black surface. In both cases, the fluid thickness was 150 μm [9] [13].
- Calculations of the efficiency of a proposed nanofluid-based receiver, appropriate for a "power tower" CSP type of powerplant (see, e.g., [27]), revealed a maximum receiver efficiency of ~75%, compared to a measured receiver efficiency of ~65% at the same temperature (300°C) [11] [28].

We thus feel there is plenty of motivation to pursue nanofluids as the basis for direct-absorption solar energy conversion. This gives rise to a number of fundamental research questions that need to be addressed for successful technology development. In the following sections, we will discuss ongoing work related to two such questions: (i) how is boiling generated in nanofluids that are exposed to concentrated light? (ii) how can the radiative properties of nanofluids be controlled to maximize light absorption and minimize losses, i.e., to optimize collector/receiver efficiency?

LIGHT-INDUCED BOILING IN NANOFLUIDS

A number of investigators have examined pool boiling in nanofluids, in which heat is added via an immersed wire or other type of heater (see, e.g., [29] [30] [31] [32]). Although conflicting results were reported in many studies, most researchers concluded that nanoparticle deposition on the heated surface played an important role. Exposing a nanofluid to concentrated light, however, enables heat—in the form of radiative energy absorbed by the nanoparticles—to be introduced into the nanofluid without a conventional immersed heater. Rather, the nanoparticles themselves, or nanoparticle aggregates, serve as the heaters (Fig. 4). This allows an investigation of boiling phenomena around a nanoscale heated surface, with potential effects arising from, say, the very small radius of curvature. At least one other study measured size effects on boiling from wires, but that investigation was limited to micron-sized wires [33]. We have not yet fully explored

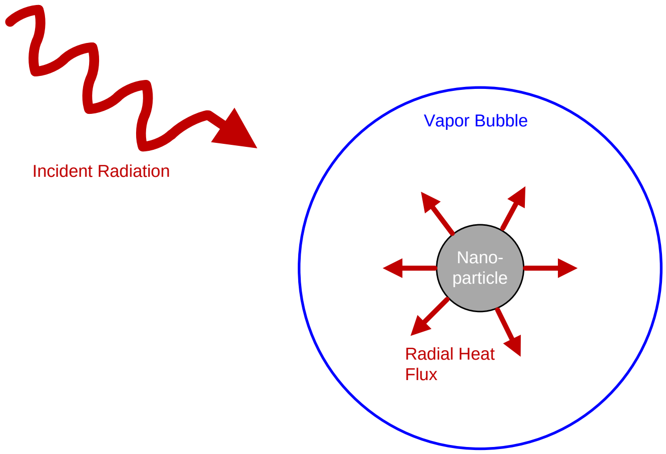

Fig. 4 Cartoon of boiling induced around a radiatively heated nanoparticle

such phenomena, but some of our results to date are discussed here.

Instead of exposing the nanofluids to concentrated sunlight, which entails a number of experimental difficulties, we chose to utilize a focused laser beam to mimic concentrated sunlight. This required only a small sample size, and allowed us to conduct experiments under controlled laboratory conditions. The experimental configuration is shown in Fig. 5. Light energy is provided by a diode pumped solid state (Coherent - DPSS) laser that gives ~120 mW at a 532nm wavelength. The laser produces a continuous column of light having a Gaussian intensity profile in the radial direction. It is focused by a 40-mm positive lens into a thin cuvette containing a 100-µm-thick fluid sample between quartz walls 1.2 and 1.3 mm thick, respectively. In the region of the sample, the focusing produces a constant diameter, 0.4-mm-beam waist which is subsequently absorbed in the nanofluid sample. In the absence of convection, the intensity of light passing though a stationary, homogeneously absorbing fluid should attenuate exponentially, according to Beer's Law. Under conditions leading to phase change in these experiments, the absorption is strong enough to absorb over 70% of the laser energy by the time it leaves the test cell, and the majority occurs in a portion of the nanofluid layer closest to the laser.

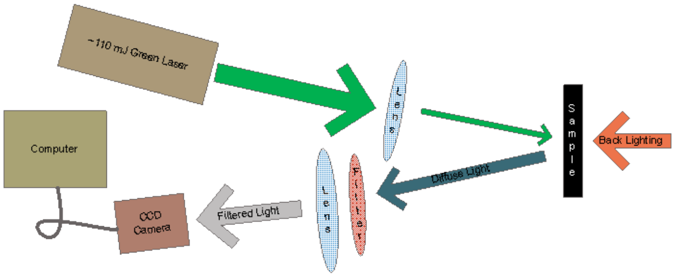

Fig. 5 Experimental configuration for laser-induced boiling in nanofluids

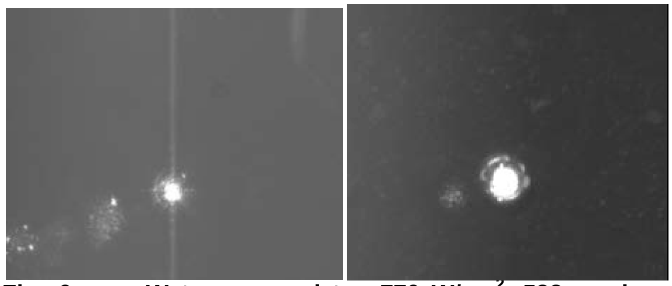

Fig. 6 Water exposed to ~770 W/cm^2, 532 nm laser light: A) in a clear cuvette, and B) in a cuvette with a black backing

The fluid behavior was observed by back-lighting the fluid layer with white light, and microscopically imaging the laser side of the cell with a Questar QM1 long range microscope (~ 3X magnification) using a Thorlabs USB 2.0, 1280 x 1024 CCD. The laser light scattered toward the microscope was removed with a low pass optical filter. Light intensity entering and exiting the cell was measured by a Coherent (FieldMaxII TOP) power meter with 0.1% accuracy.

Base Fluid Testing

The control fluid in this study is de-ionized water with ~0.1% by volume Polysorbate 80 - a surfactant. Surfactant must be used in order to create a semi-stable nanofluid. Each sample was also degassed in a vacuum chamber to remove air. Two control tests were run: a) water/surfactant in a clear cuvette, and b) water/surfactant in a cuvette with a black backing (made with 3 coats of 'satin black' Krylon spray paint on a microscope slide) attached to the exterior rear surface of the cuvette. Images of the control fluids are shown in Fig. 6.

For a clear cuvette, over 99% of the light passed through the sample – absorbing very little heat. For a black backing, the majority of light transmitted to the backing was absorbed. Heat was then conducted through the quartz of the back wall and also into the fluid. Another control fluid which absorbed light well - a black dye from Pylam Products – was also tested (not pictured in Fig. 6). This dye is composed of large-sized particles (20-800 µm as tested by a Nicomp dynamic light scattering system).

Nanofluid Testing

The tested nanofluids were copper, graphite, silver and multi-walled carbon nanotubes (nominal diameters of 2-40 nm at 1%, 0.75%, 0.5%, 0.25%, 0.1%, and 0.05% by volume) in a clear cuvette with the same DI water/surfactant base fluids. Figure 7 shows boiling induced in a φ = 0.1% graphite nanofluid heated by an 8-ns pulsed laser, while Fig. 8 presents a series of typical images during a continuous wave laser heating experiment. The lighter areas occur due to transmission of the back-lighting through a region containing a lower concentration (than average) of nanoparticles. The buoyant plume that occurs above the laser column wavers and shows vertical flow in it. The buoyancy that drives this flow may come from laser heating or from micro-bubbles emerging from the heated region. Temperatures in the plume are not likely to be high

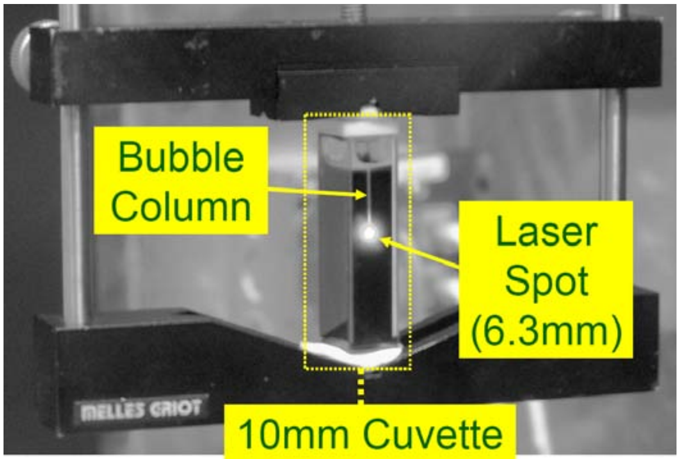

Fig. 7 **Boiling is achieved in a φ = 0.1% graphite/H_2O nanofluid by Nd:YAG pulsed laser (8 ns ~ 100mJ pulses at 15 Hz)**

enough to affect the optical density of the nanofluid, so the partial transmission associated with the buoyant plume must be due to micro-bubbles and/or particle depletion in the hot region of the laser column. The brightest regions toward the bottom of the laser beam are probably light transmitted through a vapor bubble. Thus, in this series of images (Fig. 8), a vapor bubble forms and grows in the heated region. It leaves after the laser is turned off at ~130 s. The last image shows the bubble separated from an area where the nanoparticles are heavily concentrated.

Local bubble generation in a subcooled (~ 25°C) fluid occurs only if the laser irradiance is sufficiently high. The minimum irradiance (in W/cm^2) to cause boiling in these nanofluids was found by varying laser intensity entering the fluid using neutral density filters to attenuate the beam in steps of ~100 W/cm^2. The nanofluids mentioned above—copper, graphite, silver, and multi-walled carbon nanotubes—were tested to determine the minimum irradiance necessary to initiate boiling as a function of volume fraction. Figure 9 shows trends as compared to the water/surfactant base fluid with a black backing. Some nanofluids underwent phase change for significantly less irradiance than water/surfactant with a black backing, or for water/surfactant containing Pylam black dye. Note: no vapor bubbles were observed in pure water with a clear (e.g. non-absorbing) backing for these laser fluxes – thus, this control fluid is not plotted in Fig. 9.

These experiments indicate that volumetrically heated nanofluids can undergo boiling more easily than their base fluids exposed to surface heating. In fact, up to ~50% less irradiance is necessary to create vapor in a φ = 0.75% copper nanofluid. Copper can be calculated (using the independent scattering assumption as given in [34]) to have an order-of-magnitude higher absorption efficiency than graphite at 530 nm. Since copper is the best absorber of the materials tested, it is reasonable that it will generate vapor at the lowest irradiance. This same logic can be applied to compare graphite and silver since graphite has double the absorption efficiency of silver.

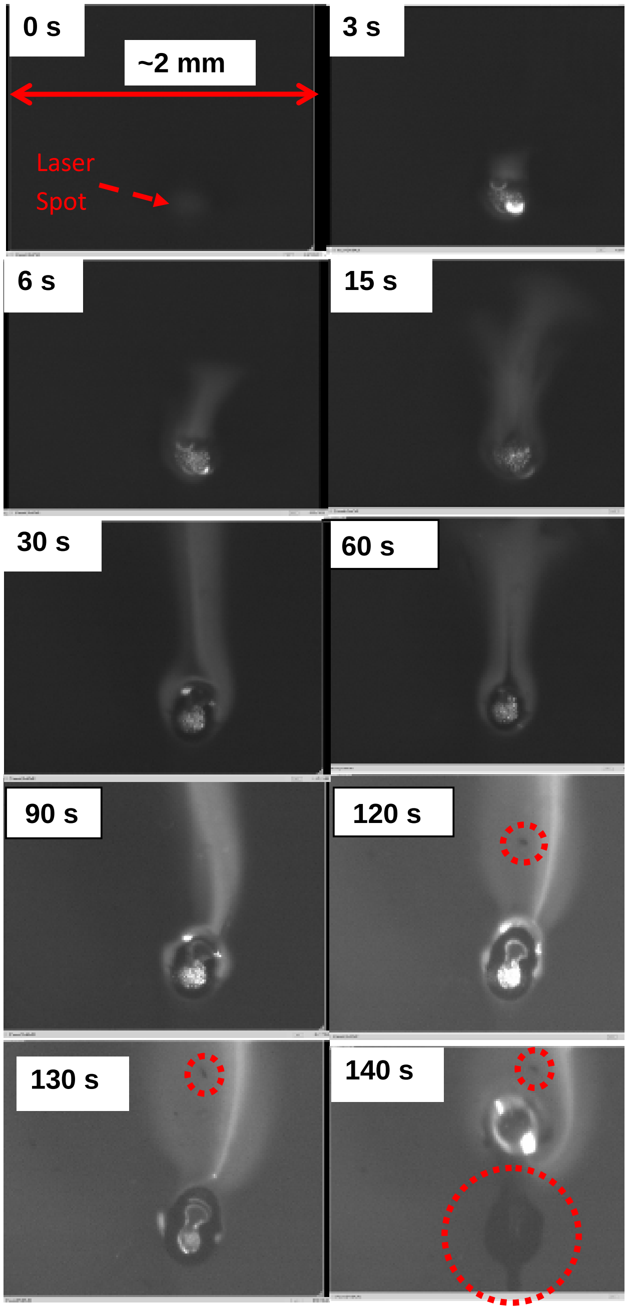

Fig. 8 **Bubble generation in a continuous-wave, laser-heated φ = 0.1% graphite/water nanofluid – dashed red circles indicate high concentrations of graphite nanoparticles**

There may be an optimum volume fraction which minimizes the irradiance necessary to drive phase change for this configuration. This is expected because low particle loadings approach the high transmittance of water and are not effectively heated. Higher particle loadings absorb the light energy close to the wall – approaching area/surface heating which may lose a significant amount of heat through the wall. The data points for copper in Fig. 9 appear to follow this trend. Trends as a

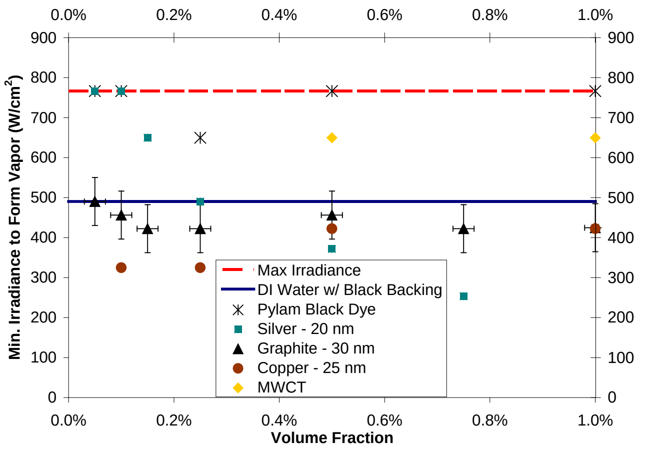

Fig. 9 **Laser irradiance needed to induce boiling. Characteristic error bars are given for graphite**

function of volume fraction for other fluids cannot be inferred from the experiments performed to date.

The experiment revealed other interesting physics that may be at work. First, phase change in pure liquid boiling commonly begins in small defects (nucleation sites) on a macroscale *surface*. For light-induced phase change in nanofluids, however, the *particles* become the heating surface. This is an important, and as yet not well-understood, difference.

Second, as indicated in Fig. 8, there are several distinct non-uniform spots in the fluid which have high concentrations of dark fluid that must be regions of concentrated particle mass. Since graphite melts at ~3,850°C and vigorous agitation can break these large regions up, it seems unlikely that these large clumps are molten and/or re-solidified graphite particles. Thus, high concentration regions are thought to be loosely bonded particle agglomerates. It is unclear whether these dark regions lead or lag vapor formation. Dense collections of particles are expected to absorb light over a shorter path length (i.e. in a smaller volume), which could cause a higher local temperature driving phase change. Conversely, as vapor forms, particles could be left behind forming high concentration regions. There is evidence for the latter in that the dark regions appear to grow with exposure time and all resolvable bubbles appear clear – i.e. lacking particles.

Third, it is unclear from the images how much vapor is leaving the heated area. Again, in macroscale boiling, vapor bubbles form, grow, and leave the surface. In Fig. 8 a bubble stays in the same spot even though buoyancy forces (calculated to be ~ 0.12 μN) should cause it to rise. Since the bubble can grow up to 500 μm in diameter in some cases, the 100-μm-thick cuvette could create a restrictive surface tension force on the bubble. Presumably, the vapor would condense in the subcooled surrounding liquid after the laser is turned off. At that point, the reduced restricting force would allow the bubble to rise. Alternatively, tiny (irresolvable) bubbles could be continuously leaving the laser spot. If so, the main bubble could be the generation site for a continuous flow of fluid in as liquid and out as microbubbles. In either case, we observe a larger final volume of vapor leave just after the laser is turned off. It should be noted that high temperature gradients should also cause particle migration away from hot regions via thermophoresis.

These results are in contrast to those predicted by recent molecular dynamics simulations [35], and by the experiments those simulations were meant to analyze [36] [37]. The experiments involved irradiation by sub-picosecond laser pulses, as compared to the nanosecond pulses and steady-state irradiation in our experiments. Those experiments, and the later molecular dynamics simulations, suggested that the heated nanoparticles could reach very high temperatures without initiating boiling. That is, a very high degree of superheat could be achieved. The results in Figs. 8 and 9 appear to contradict these earlier studies. It's possible that the differences are caused by *very* short-time-scale laser heating versus nanosecond-pulsed and steady-state laser heating, but that remains an area of study.

RADIATIVE PROPERTIES OF NANOFLUIDS

Fundamental to light-induced boiling of nanofluids is understanding of how light is absorbed by the nanoparticles, as well as by the base fluid. Here we provide a brief listing of our ongoing work in this area, and refer the interested reader to the cited publications.

- A coupled solution of the radiative transfer equation and the energy equation for a flowing nanofluid yields the temperature rise of the nanofluid as a result of absorbed radiation, and consequently the efficiency of a direct-absorption nanofluid-based solar collector or receiver [8] [9] [11] [13].
- The radiative properties were measured for four common base fluids, water, propylene glycol, ethylene glycol, and Therminol VP-1, which are likely to be utilized in direct-absorption solar thermal energy conversion systems [38].
- An analysis of radiative transfer in typical nanofluids showed that the common assumption of independent scattering cannot always be applied, but rather that multiple dependent scattering can sometimes be important [7] [10].
- The small scale of the nanoparticles leads to size effects on radiative transfer through nanofluids. This was analyzed by applying a form of the Drude model [10] [13].
- The potential exists to "tune" the spectral radiative properties of nanofluids, as represented by the extinction coefficient, through appropriate choice of nanoparticle materials, shape, volume fraction, or a combination of all these variables [12] [13].
- The volumetric radiative transfer provided by a nanofluid can lead to higher effective emissivities and/or absorptivities, relative to conventional area-based surface radiative transfer. For example, a $\varphi = 1$ %, 30-nm graphite nanofluid is calculated to exhibit a solar absorptance of ~97%, compared to ~90% for bulk graphite [12] [13].

FUTURE DIRECTIONS

Nanofluids remain a fascinating subject, as they have been for several years in the heat transfer literature and increasingly so in the solar energy literature. A number of areas for further work have already been pointed out above, but we emphasize what might be some of the most interesting subjects here:

- How can nanofluids be utilized for solar-powered direct-steam generation?
- How can the radiative properties be optimized for maximum solar energy conversion efficiency?
- What roles do enhanced convective heat transfer and thermal conductivity, as has been often observed in nanofluids, play in solar energy conversion?

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