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R. A. Taylor

P. E. Phelan

Todd Otanicar

Loyola Marymount University, totanicar@lmu.edu

R. J. Adrian

P. S. Prasher

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Vapor generation in a nanoparticle liquid suspension using a focused, continuous laser

Robert A. Taylor,^{1,a)} Patrick E. Phelan,^{1,b)} Todd Otanicar,² Ronald J. Adrian,¹ and Ravi S. Prasher¹

¹Arizona State University, School of Mechanical, Aerospace, Chemical and Materials Engineering, Tempe, Arizona 85287-6106, USA

²Loyola Marymount University, Mechanical Engineering, Los Angeles, California 90045, USA

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This letter discusses experimentation with optically induced phase change in nanoparticle liquid suspensions—commonly termed nanofluids. Four different types of nanofluids at five concentrations were exposed to a ~ 120 mW, 532 nm laser beam to determine the minimum laser flux needed to create vapor. Laser irradiance was varied between 0–770 W cm⁻². While the experiments were simple, they involved many complex, interrelated physical phenomena, including: subcooled boiling, thermal driven particle/bubble motion, nanoparticle radiative absorption/scattering, and nanoparticle clumping. Such phenomena could enable novel solar collectors in which the working fluid directly absorbs energy and undergoes phase change in a single step. © 2009 American Institute of Physics. [doi:10.1063/1.3250174]

Liquids are often used as thermal energy carriers. Many researchers have found that a small amount (<1% by volume) of nanoparticles can significantly change the thermal properties of fluids.^{1–5} A few studies have suggested that volumetrically absorbing *nanofluids*, as nanoparticle liquid suspensions are commonly called in the heat transfer literature, could eventually be used to build a highly efficient solar thermal collector.^{6–9} The operation of such a device could be affected, favorably or adversely, by the occurrence of phase change in the liquid. If solar collection is improved, some thermal energy systems (e.g., Rankine cycles, absorption cooling, water purification, ethanol distillation, etc.), which undergo phase change, might be enhanced by volumetrically absorbing nanofluids. Consequently, it may be advantageous to drive phase change with light energy in a volumetrically absorbing nanofluid, as opposed to conventional solar thermal systems in which a solid surface absorbs the sunlight and then transfers heat to the working fluid. Eliminating this additional heat transfer step makes volumetric radiative absorption potentially more efficient than conventional solar thermal collectors.

The feasibility of this concept is questioned in a recent simulation work by Merabia *et al.*¹⁰ This study concluded that curvature-induced pressure around a nanoparticle would inhibit phase change—actually melting the particle before vaporizing the base fluid.¹⁰ However, several experimental studies directed toward cancer treatments using thermal radiation have shown that *transient*, high energy laser pulses can create vapor bubbles in a nanofluid.^{11–13} Transient pulses, however, are not appropriate for solar energy harvesting applications. Therefore, this letter will present experimental measurements of nanofluids exposed to *continuous* light energy.

Light is provided by a diode pumped solid state (Coherent) laser that gives ~ 120 mW at a 532 nm wavelength. Laser light is focused by a 40 mm positive lens into a thin

cuvette containing a 100 μ m thick fluid sample between glass 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 assumed to be absorbed in the nanofluid sample according to Beer's law. The fluid behavior was observed by backlighting the fluid layer with white light, and microscopically imaging the laser side of the cell with a Questar QM1 long range microscope ($\sim 3\times$ magnification) using a Thorlabs USB 2.0, 1280 \times 1024 charge-coupled device camera. 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 (Field-MaxII TOP) power meter with 0.1% accuracy. Figure 1

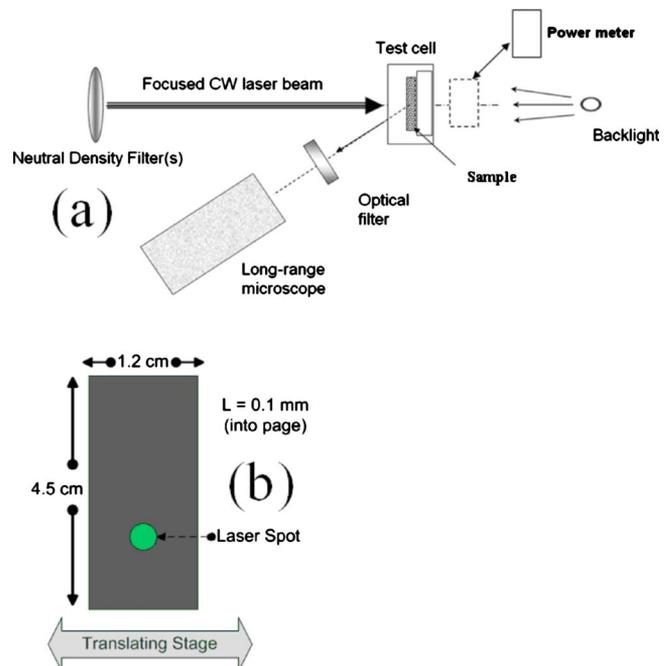


FIG. 1. (Color online) Experimental system: (a) overall test setup and (b) test cell closeup (laser beam is into page).

^{a)}Electronic mail: rataylo2@asu.edu. Tel.: (480) 727-9124.

^{b)}Electronic mail: phelan@asu.edu. Tel.: (480) 965-1625.

shows the experimental setup.

The base fluid in this study is de-ionized water with $\sim 0.1\%$ by volume Polysorbate 80—a surfactant. Each sample was degassed in a ~ 0.1 mTorr 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 three coats of “satin black” Krylon spray paint on a microscope slide) attached to the exterior rear surface of the cuvette. For (a) over 99% of the light passed through the sample. For (b) the majority of light transmitted to the backing was absorbed. Another control fluid which absorbed light well—a black dye from Pylam Products—was also tested. This dye is composed of large-sized particles ($20\text{--}800\ \mu\text{m}$ as tested by a Nicomp dynamic light scattering system).

The tested nanofluids were copper, graphite, silver, and multiwalled carbon nanotubes (nominal diameters of $2\text{--}40$ nm at 1% , 0.75% , 0.5% , 0.25% , 0.1% , and 0.05% by volume) in a clear cuvette with the same base as (a) and (b). The first (i.e., the top) two images in Fig. 2 are typical of the 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 microbubbles emerging from the heated region. Temperatures in the plume are not likely to be high enough to affect the optical density of the nanofluid, so the transmission associated with the buoyant plume must be due to microbubbles and/or particle depletion in the hot region of the laser column. It should be noted that thermophoresis is expected to cause particle migration away from the absorbing fluid which is at elevated temperature.

Local bubble generation in a subcooled ($\sim 25\ ^\circ\text{C}$) fluid is only possible if the laser irradiance is high. The minimum irradiance (in W/cm^2) necessary to cause phase change in these nanofluids was found by varying laser intensity entering the fluid using neutral density filters to attenuate the beam in steps of $\sim 100\ \text{W}/\text{cm}^2$. The nanofluids mentioned above were tested to determine the minimum irradiance necessary to cause phase change as a function of volume fraction. Figure 3 shows trends as compared to the water base fluid with a black backing. Some nanofluids underwent phase change for significantly less irradiance than water with a black backing, or for water containing Pylam black dye. Note: no vapor bubbles were observed in pure water with a clear (e.g., nonabsorbing) backing for these laser fluxes—thus, this control fluid is not plotted in Fig. 3.

These experiments indicate that volumetrically heated nanofluids can undergo liquid-vapor phase change more easily than their base fluids exposed to surface heating. In fact, up to $\sim 50\%$ less irradiance is necessary to create vapor in a 0.75% copper nanofluid. Copper can be calculated (using the independent scattering assumption¹⁴) to be the best absorber (of the materials tested) with nearly an order-of-magnitude higher absorption efficiency. It is expected that 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. 3 appear to follow this trend. Trends as a function of volume fraction

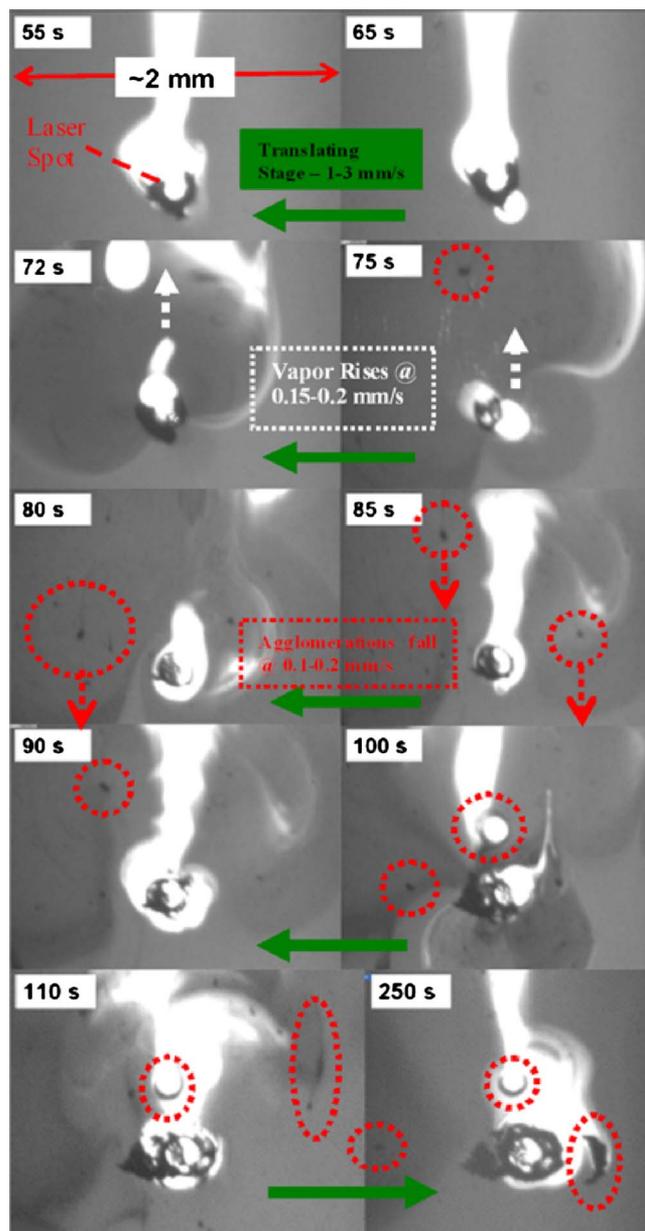


FIG. 2. (Color online) Time-lapse photos of a $0.1\%v$ graphite nanofluid exposed to a laser irradiance of $\sim 770\ \text{W}/\text{cm}^2$ —arrows indicate direction of motion via a manual translating stage at $1\text{--}3\ \text{mm/s}$ (the laser spot is in the same position in all frames). Dashed circles indicate separated high concentrations of graphite nanoparticles.

for other fluids cannot be inferred from the experiments performed to date.

As indicated in the later images of Fig. 2, there are several distinct nonuniform spots in the fluid which have high concentrations of dark fluid that must be regions of concentrated particle mass. Since graphite melts at $\sim 3850\ ^\circ\text{C}$ and vigorous agitation can break these large regions up, it seems unlikely that these large clumps are molten and/or re-solidified graphite particles as predicted by Merabia *et al.*¹⁰ 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 con-

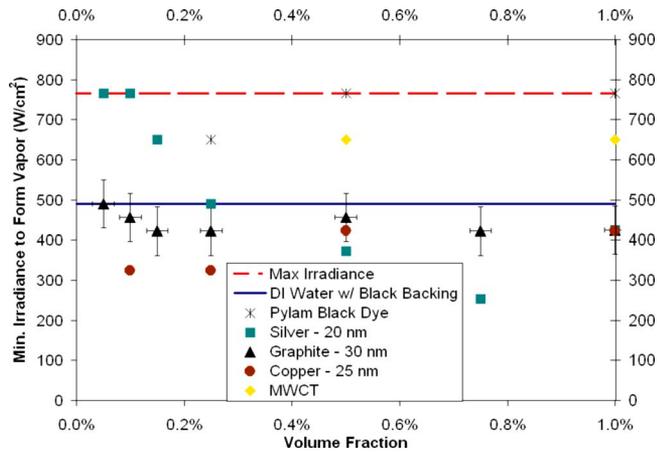


FIG. 3. (Color online) Irradiance needed to locally boil various fluids in this set-up. Characteristic error bars are given for graphite.

centration regions. There is more 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.

Additional interesting phenomena were seen in the graphite nanofluid when the laser spot was moved through the sample. The series of images in Fig. 2 show dynamic bubble expansion and contraction takes place when the sample is translated in a direction orthogonal to the incident laser beam. Since it takes some time for vapor to form in the nanofluid and particle clumps are present, some very chaotic heat and mass transfer is shown. This response was not seen in the control fluids—pure water or the black dye.

Our understanding of the complex phenomena that occur in this simple experiment is clearly incomplete, and more research will be needed to fully understand these processes.

An efficient/optimum nanofluid could be applied to many technologies that require localized heating and/or phase change. Overall, a very promising feature of nanoparticle suspensions is their versatility and controllability in optical and radiative heat transfer applications. In particular, it may be possible to develop solar collectors that absorb light directly and undergo phase change in a single step.

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