Fundamentals of Energy Transport in Nanofluids

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Scope: We performed computational simulations and theoretical analysis to investigate the underlying origins of large thermal conductivity enhancements observed in nanofluids (colloidal suspensions of solid nanoparticles and/or nanofibers in thermal fluids) and to identify strategies towards tailoring nanofluids for better thermal performance.

Background: Over the last decade nanofluids sparked excitement as well as controversy. In particular, a number of researches reported dramatic increases of thermal conductivity with small nanoparticle loading, while others showed moderate increases consistent with the effective medium theories on well-dispersed conductive spheres. Accordingly, the mechanism of thermal conductivity enhancement was a hotly debated topic. Without understanding of the mechanism our ability to tailor and predict performance of nanofluids in limited to empirical observation thus limiting application of nanofluids to engineering systems..

Major Achievements:

Understanding of the Mechanism: In our first publication on nanofluids we opened a discussion on possible mechanisms responsible for unusually high thermal transport properties of nanofluids [1], including effects of liquid layering at solid interface, Brownian motion and particle clustering. This paper is now cited 200 times (scopus.com), the most of any theoretical article on the topic. In subsequent publications we examined in detail various effects, including liquid layering [2,3], Brownian motion, [4] and particle clustering [5, 6]. Findings of our research combined with the analysis of the experimental data clearly demonstrated that the only mechanism capable of explaining thermal conductivity of nanofluids is particle clustering. Furthermore, the vast majority of the published experimental data can be explained by the classical effective medium theories that account for particle aggregation [7] without resorting to novel mechanisms such as Brownian motion induced nanoconvection. This finding has important practical implication: Particle aggregation required to significantly enhance thermal conductivity also increases fluid viscosity rendering the benefit of nanofluids to flow based cooling applications questionable.

Carbon Nanotube Suspensions: We studied the problem of the heat flow between organic liquids and carbon nanotubes. This work was in collaboration with Prof. David Cahill from University of Illinois at Urbana-Champaign, who was also supported by the DOE (Grant No. DEFG02-91-ER45439). In this work we found that the tube-liquid interfacial resistance is very high due to weak coupling between high frequency thermal vibrations in tubes with soft (low frequency) vibrations in the liquid [8]. This result provides an explanation for an order of magnitude lower than expected thermal conductivity enhancement observed for carbon nanotube based suspensions and polymer nanocomposites, and suggest that further progress in the composite thermal conductivity enhancement will require tailoring interfacial bonding and increasing the aspect ratio of carbon nanotubes [9]. The importance of our results was recognized by a publication in the Nature Materials Journal [8], which is already cited well over 100 times (scopus.com), and made a cover of Materials Today (Fig. 1). We also explained why despite very low percolation thresholds exhibited by carbon nanotube polymer composites signified by steep increases in electrical conductivity at very low tube loadings there are no signatures of percolation in thermal transport. In particular, we performed a theoretical analysis based on finite element calculations [10] that exposed the key reasons for markedly different behaviors of electrical and thermal transport: (i) Much larger fiber to matrix electrical conductivity ratios, that the corresponding ratios for thermal conductivities, and (ii) fiber-matrix and fiber-fiber interfacial resistances. Our findings suggest that rather than concerning oneself with the percolation problem in development of more thermally conductive nanofiber composites one should focus on increasing fiber aspect ratio and decreasing interfacial thermal resistance.

Fig. 1: Cover of the June 2005 Materials Today with a nanotube-organic matrix structure form our simulations.

Nanoscale Thermal Effects Relevant to Biological Systems: Based on an analysis of the diffusive heat flow equation, we determined limits on the localization of heating of soft materials and biological tissues by electromagnetically excited nanoparticles.[11] For heating by rf magnetic fields or heating by typical continuous wave lasers, the local temperature rise adjacent individual nanoparticles is negligible. Only heat dissipation for a large number of nanoparticles dispersed in a macroscopic region of a material or tissue produces a global temperature rise that is capable of affecting biofunctionality. This finding implies that individual continues nanoscale heat sources cannot be used to thermally control the biofunctionality or cause hypothermia.

Training: Over its entire length this program partially supported three graduate students, Liping Xu (Ph. D 2004), William Evans (Ph.D. 2008) and Natalia Shenogina (expected Ph.D. 2009). All of the students acquired in depth knowledge of molecular and continuum level modeling of heat flow in nanoscale systems and at various material interfaces. Currently Liping Xu works as a researcher at University of Utah University and William Evans works at Knolls Atomic Power

Laboratory at Niskayuna, both contributing to US economy and its global competitiveness. The third student (Natalia Shenogina) is not yet graduated.

Summary and Impact: Support under DOE contract resulted in development of comprehensive understanding of the mechanism of the enhanced heat conduction in nanofluids relying on the clustering of particles and thus creation of extended paths for heat flow along highly conductive particle clusters. In course of our research we, along with other nanofluid researchers, contributed to disqualification of a number of "new" mechanisms and thus focus the field on the relevant issue, i.e., the cluster structure of nanoparticle suspensions. Our work on nanotube nanofluids and polymer composites identified interfacial resistance as a major obstacle for the effective utilization of superior thermal transport properties of carbon nanotubes. Our work has a major impact on the field of nanofluids and nanocomposites demonstrated by 20 publications with 2 already cited over 100 times, numerous invited presentations in the US and abroad, invited articles, and magazine covers. Our most important practical message is that in tailoring thermal transport of nancomposites and nanofluids the key factors are particle clustering and interfacial transport properties.

Cited publications resulted from the contract support.

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Other publications resulted for the support of the contract.

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