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Enhanced thermal transport through a soft glassy nanodisk paste

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We study the diffusion of heat in an aqueous suspension of disk-shaped nanoparticles of Laponite, which has finite elasticity and a pastelike consistency, by using the Mach-Zehnder interferometer. We estimate the thermal diffusivity of the suspension by comparing the experimentally obtained temperature distribution to that with an analytical solution. We observe that, despite the highly constrained Brownian diffusivity of particles owing to its soft glassy nature, suspensions at very small concentrations of Laponite demonstrate significant enhancement in thermal diffusivity. We correlate the observed enhancement with the possible microstructures of the Laponite suspension.

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I. INTRODUCTION

With advancing technologies and continuously shrinking devices, such as electronic gadgets, chemical (micro) reactors, and fuel cells, there is an ever increasing demand for a medium with enhanced thermal transport properties. Heat can, in principle, be transferred by three mechanisms: conduction, convection, and radiation [1]. However, at miniature scales, small gaps, and moderate temperature gradients, conduction is the dominant mode of heat transfer. An important requirement for effective heat transfer to the environment in media, such as heat sink pads, is close contact between the sink and the instrument without gaps or air pockets. It necessitates the material to be easily deformable and to be strong enough to support the weight of the gadget. In this paper, we study the heat transport characteristics of a soft glassy solid with a pastelike consistency. It comprises an aqueous suspension of nanodisks of Laponite, a model soft glassy material [2]. We believe that this system is unique due to the oblate (disklike) anisotropic shape of the suspended particles, whose heat transport behavior has scarcely been studied in the literature. This soft solidlike suspension is observed to demonstrate remarkable enhancement in thermal diffusivity at very low volume fractions.

In order to enhance performance, a single phase material is routinely mixed with a small percentage of additive or filler, having significantly different physical properties. If both the major and the minor components are in the solid state, the resultant combination is represented as a composite. If the major component is liquid and the minor component is solid particles, we get a suspension which retains the viscous properties of the liquid in the limit of a small fraction of solid particles. In both situations, if the physical dimension of the solid particles is in nanometric range, the former is called a nanocomposite, whereas, recent literature describes the latter as a nanofluid. With regard to heat transfer behavior, suspensions including nanofluids can transport heat by conduction as well as convection mechanisms. However, owing to their fluid character, they do not possess enough physical strength necessary to support heat producing gadgets. Composites, on the other hand, can transport heat only by conduction. In addition, composites do possess the necessary strength to support weight but cannot provide a very close contact without gaps.

Suspensions of particles having sizes in the micrometer range are observed to follow the Maxwell relation that relates the effective thermal conductivity to its volume fraction (ϕ). This relation suggests enhancement to scale as 3ϕ when particle thermal conductivity is much larger than the suspending liquid conductivity [3–5]. However, the recent literature suggests that, compared to micrometer sized particles, suspensions of nanoparticles, namely, nanofluids, show significant enhancement in thermal conductivity when compared at equal volume fractions [6,7]. Typically, an enhancement of 10%–120% for a particle volume fraction of 2% is reported in the literature for various nanofluids [3,4,6], whereas, the Maxwell relation predicts a maximum enhancement of only 6%. Various mechanisms proposed to explain this unusual behavior include phonon transport [6], Brownian diffusion [8], molecular ordering on nanoparticles [9], and cluster or aggregate formation [10–12]. However, most of these mechanisms cannot completely explain the anomalous increase in thermal conductivity available in the reported experimental data. On the other hand, there is a broad consensus in recent literature on the possible role of aggregate formation in the observed increase in thermal conductivity [3,6].

Heat transport of both the microcomposites as well as the nanocomposites has been studied in great detail [13]. Typically, the conductivity of the suspended medium, extent of aggregation, its aspect ratio and orientation, quality of interface between suspended particles, and the density of percolation in the matrix determines the heat transport behavior [14–18]. However, contrary to that observed in nanofluids, some papers suggest that thermal conductivity of nanocomposites is lower than that seen in microcomposites for a given loading of the filler [16]. Such a phenomenon is attributed to the enhanced

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resistance associated with the particle—matrix interface and is expected to have a pronounced effect at nanoscopic dimensions.

In this paper, we study the heat transport behavior of an aqueous suspension of disklike nanoparticles of Laponite RD, which has soft solidlike consistency and, therefore, is intermediate to nanofluids and nanocomposites. Laponite RD is a synthetic silicate clay mineral, having a disklike shape with a diameter of 25 nm and a thickness of 1 nm [19]. Although not available in the literature, thermal conductivity of a Laponite particle is expected to be higher than water, owing to its structure as a single crystal [20]. In an aqueous medium with a pH of 10, the Laponite disk is known to possess a negative charge on opposite faces with a weakly positive charge on its edge. The addition of salt screens the negative charge, thereby, enhancing attraction among Laponite particles. Overall, a combination of attractive and repulsive electrostatic forces leads to complex interaction among the particles [21–24] so that incorporation of Laponite in water increases its viscosity and elasticity by over several orders of magnitude, eventually rendering a soft solidlike consistency [2,25,26]. Laponite suspension is also known to possess yield stress [27]. Several scattering studies have shown that such massive increases in viscosity and elasticity are due to the caging of particles, causing a strong reduction in Brownian diffusivity of disk-shaped nanoparticles [28,29]. Consequently, Laponite suspension undergoes egrodicity breaking and owing to its pasty nature, it is represented as a soft glass [29].

II. EXPERIMENTAL PROCEDURE AND MATERIAL PREPARATION

In the present paper, we estimate the thermal diffusivity of a soft glassy aqueous Laponite suspension using laser interferometry through an unsteady heat diffusion experiment. In a typical experiment as shown by a schematic in Fig. 1, a slab of material of thickness H and thermal diffusivity α is maintained at temperature T_C. At time $t = 0$, the temperature of the top plate is suddenly raised to T_H. The thermal field follows a diffusion equation $\partial\theta/\partial\tau = \partial^2\theta/\partial\eta^2$, where $\theta = (T - T_C)/(T_H - T_C)$, $\tau = \alpha t/H^2$, and $\eta = y/H$ [30]. For a constant α, this diffusion equation can be analytically solved for known initial and boundary conditions ($\theta = 0$ at $\eta = 0$; $\tau \leqslant 0$, $\theta = 0\,\forall\,\eta$; $\tau > 0$, $\theta = 1$ at $\eta = 1$) to yield the time evolution of the temperature field as

$$\theta = \eta - 2\sum\nolimits_n [(-1)^{n+1}/n\pi]\sin(n\pi\eta)\exp(-n^2\pi^2\tau),\quad n = 1,2,3,\ldots, \tag{1}$$

By comparing experimental results with Eq. (1), parameter α can be estimated.

We procured Laponite RD® from Southern Clay Products, Inc. The suspension of Laponite in ultrapure water is highly transparent. Suspension was prepared in water having a pH of 10 and a predetermined amount of salt (NaCl) using the procedure described elsewhere [23]. A freshly prepared suspension was stored in a sealed polypropylene bottle for around 30–50 days. Before carrying out the interferometry experiment, an aged suspension was vigorously stirred. A presheared Laponite suspension was poured in the test chamber and was left undisturbed for around 4 h at constant temperature T_C. In this paper, we have used the Mach-Zehnder interferometer, wherein the split collimated laser beam [632.8 nm, 35 mW He-Ne laser (Spectra-Physics)] is passed through the test and the reference chambers. The two beams are superimposed before being imaged by a CCD camera as shown in the schematic in Fig. 2. The optical path difference, created during the experiment between the reference and the test beams, produces an interference pattern. The reference chamber was filled with glucose solution. Before beginning the experiment, the refractive index of the glucose solution was matched with the sample in the test section, ensuring no optical path difference between the two light beams. Subsequently, the temperature of the top plate was increased to T_H. Temperatures adopted in various experiments were $T_C = 24°\text{C}–28°\text{C}$ and $T_H = 27°\text{C}–30°\text{C}$ (the bottom plate temperature, maintained using a constant temperature bath, was usually kept equal to the ambient temperature which varied between 24°C and 28°C). The hot and cold surfaces were held at spatially and temporally uniform temperatures by constant temperature baths. As time passes, heat diffuses from the heated top plate towards the bottom increasing temperature within the medium. The change in temperature alters the refractive index of the suspension creating an optical path difference between the test and the reference chambers, thereby, producing an interference pattern.

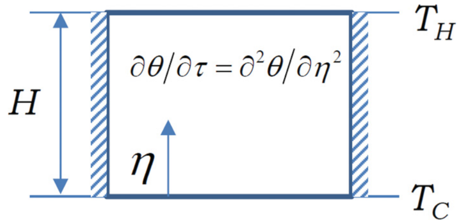

FIG. 1. (Color online) Representation of a thermal field. A slab of Laponite suspension having thickness H is maintained at temperature T_C for $t \leqslant 0$. For $t > 0$, the temperature of the top surface is increased to T_H.

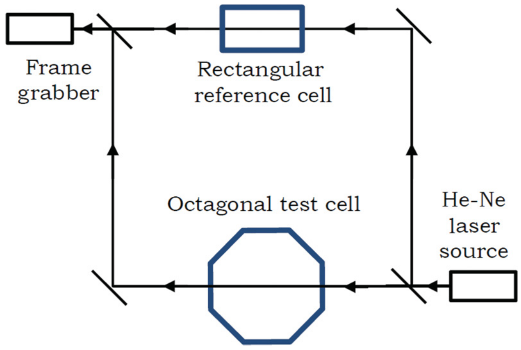

FIG. 2. (Color online) Schematic of a Mach-Zehnder interferometer. The test cell has an octagonal cross section. This cell is a top view of the arrangement shown in Fig. 1. The test beam and reference beam meet to produce an interference pattern, depending upon the path difference between the two.

Aqueous suspension of Laponite is known to demonstrate thixotropy, leading to time and deformation field dependent viscosity and elasticity [2,27]. Under such conditions, elastic and viscous moduli give a reliable measure of the rheological behavior of the sample. We measured elastic and viscous moduli of a 30 day old sample. In a typical experiment, we applied an oscillatory shear stress ($\sigma = \sigma_0 e^{i\omega t}$) having magnitude $\sigma_0 = 0.5$ Pa and frequency $\omega = 0.1$ Hz to the samples 4 h after shear melting the same in a rheometer. The experiments were carried out at 25°C using a MCR 501 rheometer. Owing to the applied stress field, strain induced in the material has a form $\gamma = \gamma_0 e^{i(\omega t - \delta)}$. The elastic ($G'$) and viscous ($G''$) moduli are given by $G' + iG'' = \sigma_0 e^{i\delta}/\gamma_0$ [31]. The heat capacity of dry Laponite powder was also measured using a differential scanning calorimeter and was found to be 1.03 kJ kg^{-1} K^{-1}.

III. RESULTS AND DISCUSSION

Subsequent to an increase in temperature of the top plate, as a step function, heat diffusion towards the lower plate gradually increases the suspension temperature, leading to an optical path difference between the test and the reference chambers. In Fig. 3, we describe the corresponding evolution of the interference pattern. Due to the diffusion of heat from the top surface, fringes appear in its vicinity and migrate towards the lower surface with time. In the limit of long times, a steady state sets in, and fringes get uniformly distributed. The temperature difference between two consecutive fringes (ΔT_ε) can be estimated from the principles of wave optics and is given by $\Delta T_\varepsilon = \lambda/(L\, dn/dT)$, where λ is the wavelength of light, L is length of the test cell, and dn/dT is the variation in the refractive index with temperature [32]. We used values of dn/dT for Laponite suspension available in the literature [33]. Therefore, with the knowledge of temperature at one boundary and the location of the fringes, the complete thermal field can be obtained [34]. In Fig. 4, we plot the evolution of temperature as a function of time from the interference pattern shown in Fig. 3. These results are essentially equivalent to the analytical solution [Eq. (1)] for various time instants. At short times, the temperature data are not very reliable due to sharp gradients that cause refraction of the light beam. In addition, in the limit of short and long times, the temperature field is not sensitive to the value of α. Closer to the steady state, the temperature field in the suspension is affected by the thermal properties of the confining surfaces. Quantitatively, the sensitivity of the estimated parameter to the measurement is expressed by $S(t,\alpha) = \int_0^H (dT/d\alpha)dy$ [35]. In order to estimate α, we fit Eq. (1) to the experimental data obtained in that window, where $S(t,\alpha)$ is high, by a procedure that is equivalent to the method of weighted least squares. In Fig. 4, the fit of the analytical solution (displayed by lines) is shown to only those data which are contained in the 90% sensitivity window. It can be seen that the data fit the analytical solution very well. It should be noted that the height of the test cell is $H = 50$ mm and the sample is stably stratified in the density. Hence, there are no convection currents, and the present method measures the bulk thermal diffusivity α of the solution.

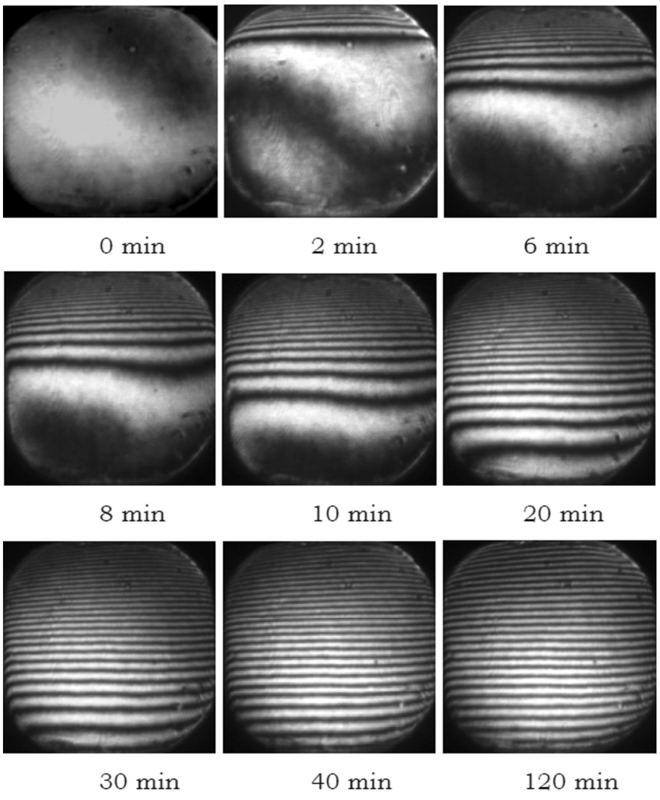
0 min 2 min 6 min
8 min 10 min 20 min
30 min 40 min 120 min

FIG. 3. Evolution of the interference pattern as a function of time for a 0.8 vol % aqueous Laponite suspension. The upper and lower surfaces were maintained at 27°C and 25°C, respectively.

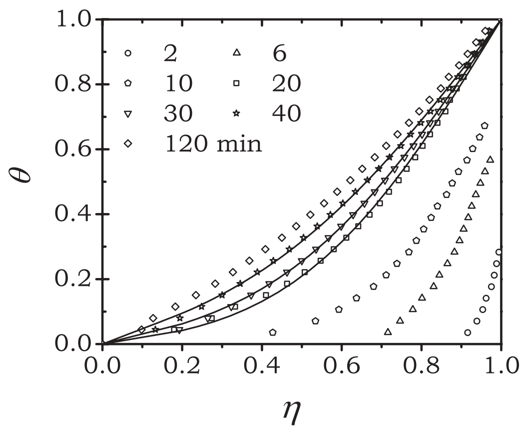

FIG. 4. Evolution of the dimensionless temperature as a function of the dimensionless distance from the bottom plate at various times for interference patterns shown in Fig. 3. The symbols are the experimental data, whereas, the lines are the fits of Eq. (1) to the experimental data.

In Fig. 5, we plot α as a function of Laponite concentration (ϕ_L). For pure water ($\phi_L = 0$), the analysis of interferometry data estimates the value of α very close to that given in the literature ($\approx 1.4 \times 10^{-7}$ m^2/s). It can be seen that the incorporation of Laponite in water leads to considerable enhancement in α. In the inset of Fig. 5, we plot the effect of the concentration of NaCl (C_s) on α for 0.8% and 1% Laponite concentration suspensions. It can be seen that, within the experimental uncertainty, α changes weakly with changes in C_s.

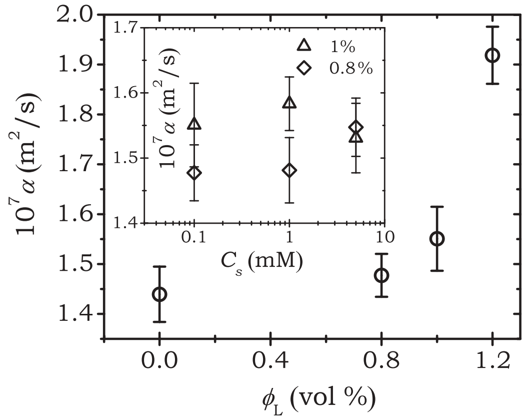

FIG. 5. Thermal diffusivity of the aqueous Laponite suspension is plotted as a function of vol % of Laponite (ϕ_L) without any externally added salt ($C_S = 0.1$ mM). The inset shows the effect of the concentration of Na$^+$ ions (C_S) on the thermal diffusivity of the aqueous suspension of Laponite for $\phi_L = 0.8$ and 1 vol %.

We also estimate the thermal conductivity ($k = \rho C_P \alpha$) of the Laponite suspension from the experimentally determined heat diffusivity data (Fig. 5). Heat capacity of the aqueous suspension is obtained by using the rule of mixtures $C_{P,mix} = \sum_i w_i C_{Pi}$, where w_i and C_{Pi} are the mass fraction and the heat capacity of the ith fraction, respectively. In Fig. 6, k for the Laponite suspension is plotted as a function of ϕ_L, whereas, in the inset, we plot the same as a function of C_s. It can be seen that the data follow the same trend as the thermal diffusivity shown in Fig. 5.

We also carried out oscillatory rheological experiments on the suspension samples. As mentioned in the Introduction, the aqueous suspension of Laponite for the concentration range (of Laponite as well as salt), explored in this paper, has a soft solidlike consistency. In Fig. 7, we plot elastic and viscous moduli of the aqueous suspension of Laponite as a function of the concentration of Laponite. Irrespective of the concentration of Laponite in the explored range, we observe the elastic modulus to always be greater than the viscous modulus ($G' \gg G''$). It has been observed that the elastic modulus of the aqueous suspension of Laponite is a very weak function of frequency over the practically explorable frequency range and, therefore, should be termed as a soft solid (with infinite viscosity) [36]. Such rheological behavior also indicates the significantly constrained translational diffusivity of the Laponite particles in the suspension. Similar to the sample without salt, samples having salt also show qualitatively similar rheological behavior (not shown in the figure) with a soft solidlike nature [27]. The results of Figs. 5–7 are striking as significant enhancement in α is observed despite constrained Brownian diffusivity [28,29].

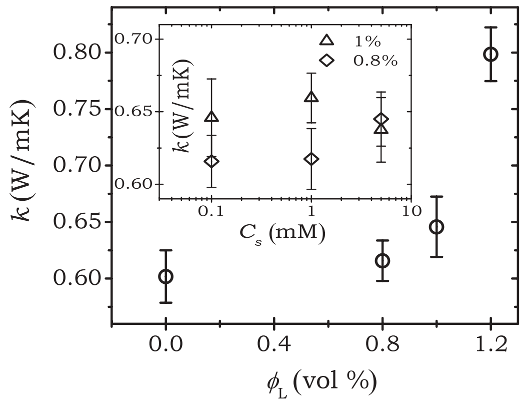

FIG. 6. Thermal conductivity of the aqueous Laponite suspension is plotted as a function of vol % of Laponite (ϕ_L) for the data shown in Fig. 5. The inset shows the effect C_S on the thermal conductivity of the aqueous suspension of Laponite for $\phi_L = 0.8$ and 1 vol % for the data shown in Fig. 5.

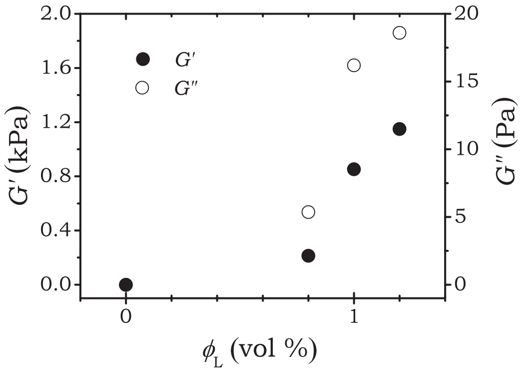

FIG. 7. Filled circles: elastic modulus and open circles: viscous modulus of the aqueous Laponite suspension are plotted as a function of vol % of Laponite (ϕ_L).

The microstructure of the Laponite suspension which restricts Brownian diffusivity remains a matter of discussion [21,22,36,37]. Some groups have proposed that the aqueous suspension of Laponite typically above 0.8–1 vol % (2–2.5 wt%), forms a repulsive glass [21,24]. In this state, the Laponite particles, owing to electrostatic repulsion among them, remain in a self suspended state (without touching each other). On the other hand, some groups believe the aqueous suspension of Laponite is a gel formed by an interconnected network of fractal aggregates of particles [22], which form a percolation path. The precise microstructure responsible for enhanced thermal transport in the present system, whether a repulsive glass or an attractive gel, is the subject of continuing discussions [36,38]. We postulate the following reason for the enhancement of heat transport in the Laponite suspension, viewed from independent perspectives: as glass as well as gel.

In case the Laponite suspension forms a repulsive glass wherein particles do not touch each other, heat transport can occur primarily via transfer of the thermal motion. Typically, for 0.8, 1, and 1.2 vol % Laponite suspensions, average interparticle distances are equal to 40, 37, and 34 nm, respectively, the average interparticle distance decreasing as $\sim\sqrt[3]{\phi_L}$ [21]. However, owing to the anisotropic shape of the particle (a disk with 25–30 nm diameter and 1 nm thickness), the nearest points can be significantly closer than the average value. The Laponite particle, due to its crystalline nature, is expected to have higher thermal conductivity than water. Therefore, the transfer of heat can get enhanced through the particles in the suspension. A decrease in distance between the particles caused by an increase in concentration is anticipated to enhance heat transport further as shown in Figs. 5 and 6.

When Laponite suspension is visualized to form a network of fractal aggregates, heat transport can be seen as occurring on two scales: one, the aggregates that fill the space and the other, within the aggregate where Laponite disks form a fractal network. In this proposal, Laponite particles touch each other in an edge-to-face fashion. Hence, the transfer of heat can take place more effectively through the percolation network. In this case, the volume fraction of aggregates ϕ_a, which is also indicative of the density of the percolation network, is related to ϕ_L as $\phi_L = \phi_i \phi_a$ [11], where ϕ_i is the volume fraction of the Laponite particles within an aggregate. An increase in ϕ_i enhances the thermal conductivity of the aggregate, although at the expense of the density of the percolation network. Therefore, this picture requires a fine balance between ϕ_i and ϕ_a, so as to cause enhancement in overall heat transport. According to Mongondry *et al.* [22], density of a three dimensional network increases with Laponite concentration, thereby, making more paths per unit area for the heat transport available.

As mentioned before, nanofluids were also observed to demonstrate significant enhancement in thermal conductivity compared to the base fluid. The recent literature on nanofluids suggested the important role played by fractal aggregates in enhancing its thermal conductivity [3,6]. It should be noted that the rheological constitution of nanofluids with respect to that of the present system is completely different. The nanofluids are essentially in liquid states, have finite viscosities, and have no measurable elasticity. The aqueous suspension of Laponite, on the other hand, has infinite viscosity (in the limit of weak deformation fields) and finite elasticity with a pasty or soft solidlike consistency. Additionally, owing to the formation of aggregates, sedimentation may cause the nanofluids to become unstable [39,40]. On the other hand, sedimentation is not observed in Laponite suspension beyond the concentration of 0.4 vol % [24].

The present paper is unique from various points of view. First, very little work is available in the literature, experimental as well as theoretical, regarding heat transport through the suspension of oblate (disklike) particles. The present system has a consistency of a paste and is described in the recent literature as soft glass [2]. Very little work has been performed on such systems to study their heat transport characteristics. Additionally, interferometry as a technique, which determines the properties of materials in bulk, is being used to estimate heat transport parameters in suspensions. In the present paper, we report remarkably enhanced bulk heat transport in a pastelike medium at very small concentrations of the suspended medium. We believe that the results mentioned in this paper will attract new applications for pasty materials and will stimulate further discussions.

IV. CONCLUSION

In this paper, we investigated conduction heat transfer in the pasty suspension of nanodisks of Laponite in water using a Mach-Zehnder interferometer. The elastic modulus of Laponite suspension was observed to be significantly greater than the viscous modulus, indicating the material to be in a solid state with highly constrained Brownian diffusivity of particles in the suspension. In the heat transfer experiments, transient evolution of temperature as a function of space was obtained using interferometry. The thermal diffusivity of the suspension was obtained by comparing the experimentally obtained time dependent temperature profile with the analytical expression. We observed that thermal diffusivity increased with the concentration of Laponite. Remarkably, the material showed significant enhancement for very small concentrations of Laponite.

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