Sedimentation in nanofluids during a natural convection experiment

K. Kouloulias*, A. Sergis, Y. Hardalupas
Imperial College London, Mechanical Engineering Department, London SW7 2AZ, UK

1. Introduction

High performance cooling is one of the most vital needs of many industrial technologies to date, from micro/nanoelectronic mechanical systems (MEMS/NEMS) to nuclear reactors. Material science and technology has reached its limits regarding the increase of the solid-fluid interface area. In addition, downsizing appears to be the current trend. As a result, high heat fluxes have to be accommodated to ensure a safe, efficient and long-lasting operation of the heat exchangers and subsequently of the systems involved. As a consequence, there is a variety of engineering applications that demand the development of new coolants with superior thermophysical characteristics. One such application is fusion reactors in the nuclear energy sector, where extremely high heat fluxes (up to 20–30 MW/m²) are present in both the current generation of experimental reactors, as well as future Fusion power plants [1].

Recent advances in nanotechnology have allowed the development of a new category of coolants, namely nanofluids, as initially coined and proposed by Stephen U.S. Choi of Argonne National Laboratory in the early 1990s, while the first study was carried out in 1995 [2,3]. By definition, nanofluids are a new class of heat transfer fluids engineered by dispersing and stably suspending nanoparticles with typical size of the order of 1–100 nm in traditional heat transfer fluids [4]. Common materials used for nanoparticles

* Corresponding author.
E-mail address: k.kouloulias13@imperial.ac.uk (K. Kouloulias).
Natural convection as a heat transfer mechanism is widely applied to a variety of applications, where the thermal management results only from the density difference of the working fluid involved. However, limited experimental studies have been carried out on nanofluid natural convection, compared to conduction and forced convection. While substantial enhancement of heat transfer is reported for forced convection, contradictory results are present between numerical and experimental studies for natural convection in the literature. In most of the theoretical or numerical works, heat transfer enhancement is reported, whereas in experimental investigations unexpected deterioration is observed. Khanafar et al. [16] investigated numerically natural convection of copper-based nanofluids in a two-dimensional enclosure. Their results suggested substantial increase of the heat transfer rate for any given Nusselt number. In addition, as the nanoparticle volume fraction increased, the heat transfer ratio also increased. Ozturk and Abu-Nada [17] modeled natural convection of different nanofluids in a partially heated enclosure. They reported that the mean Nusselt number increased with the volume fraction of nanoparticles over the whole Rayleigh number range. Also, their results indicated the importance of both heater location and dimension, as well as the aspect ratio of the enclosure on heat transfer enhancement, when using nanofluids. Aminossadati and Ghasemi [18] performed a numerical study for natural convection of various nanofluids in a partially heated enclosure. They indicated that Al2O3, TiO2 and even Cu and Ag nanoparticles provided high cooling performance by reducing the maximum temperature of the heat source significantly. Meng and Li [19] investigated numerically Al2O3–H2O nanofluids under natural convection in a horizontal cylinder. Their work revealed that the nanofluid did not exhibit improved heat transfer performance than the base fluid. In addition, as the nanoparticle concentration increased, the heat transfer deterioration became more predominant. Putra et al. [9] used Al2O3 and CuO nanoparticles suspended in water. Their experimental results indicated that the natural convective heat transfer of nanofluids was lower than that of pure water and that the deterioration increased with increased particle concentration. Also, the deterioration was greater for CuO than for Al2O3 nanofluids. This difference was attributed to the slip mechanism between the denser CuO particles, compared to Al2O3 particles, and the carrier fluid. Both the particle–fluid slip and the sedimentation mechanisms appeared to be of great importance for the heat transfer performance of the nanofluids. Wen and Ding [20] studied experimentally the performance of TiO2–H2O nanofluids under natural convection. They confirmed that deterioration of natural convective heat transfer increased with nanoparticle concentration. Possible explanations for this behavior were the convection induced by concentration difference, the particle–surface and particle–particle interactions and the modification of the dispersion properties. Nnanna [21] reported different experimentally measured convective heat transfer performance for small and large nanoparticle volumetric fractions, φ, for Al2O3–H2O nanofluids under natural convection. For 0.2% < φ < 2.0%, the natural convective heat transfer was enhanced, whereas for φ > 2% heat transfer deterioration was reported. The augmentation was credited to the thermo-hydrodynamic effect that is caused by the changes in thermal conductivity and viscosity, where deterioration was attributed to the reduction of the Rayleigh number due to the increased kinematic viscosity. Li and Peterson [22] investigated experimentally the heat transfer characteristics of Al2O3–H2O nanofluids. They reported a systematic deterioration of heat transfer with an increase of the volume fraction of nanoparticles. The observed performance was mainly attributed to the higher viscosity of nanofluids, the Brownian motion and thermophoretic motion.
of nanoparticles. Ni et al. [2] conducted fully turbulent natural convection experiments with water-based alumina nanofluids. Their results indicate that the convective heat transfer coefficient, the Nusselt and the Rayleigh numbers all decrease with increasing nanoparticle volumetric concentrations between 0% and 1.08%. The mass diffusion of the nanoparticles inside the nanofluid was suggested as a possible reason for the measured deterioration.

In cases where stationary or buoyancy-driven nanofluids are involved, the physical characteristics and the long-term stability of the nanoparticle suspension play key roles. The stability of an aqueous nanofluid is directly linked to its electrokinetic properties as high surface charge results in strong repulsive forces between the nanoparticles [23]. In the literature there are many effective methods for the preparation of stable suspensions. The most widely applied methods are the ultrasonic vibration, the electrostatic stabilisation and the steric stabilisation [23,24]. Numerous studies have been conducted on the analysis of nanofluid stability pointing out the critical effect of the pH of the carrier fluid [25–28].

The overall behavior of a nanofluid depends on whether the water is acidic or alkaline. The surface of Al2O3 nanoparticles in acidic and neutral DI water has a positive charge, whereas in alkali water it is negatively charged [26,29]. However, there is a certain pH value where a nanofluid reaches an equipotential point, the isoelectric point (IEP). At this point, the electric charge on the particles is zero [23,30]. As a result, the ideal pH corresponds to a high surface charge or zeta potential [27,31]. An increased zeta potential corresponds to stronger electrostatic repulsion forces among the charged nanoparticles due to the electric double layer that is formed around the particles. It is well studied that a suspension with zeta potential below 20 mV exhibits limited stability, whereas for 30 mV and above, it is characterized as physically stable [29]. However, the IEP depends not only on the ionic constitution of the base fluid, but also on the material, phase and purity of the nanoparticles. For γ-Al2O3, the IEP is near 7.7–7.9 [31]. Wen and Ding [28] concluded that for γ-Al2O3 a pH of 7 is appropriate for their experimental work, in the sense that it was reasonably away from the IEP and no damage would be caused to their heating surface. Pak and Cho reported that a pH value of 3 would ensure good homogeneous dispersion, whereas Adegbite [31] calculated the optimum pH value at 4. Mao et al [25] noted that when the pH was less than 7, the stability was improved, but for higher values sedimentation appeared in the system.

Therefore, the literature shows that the heat transfer behavior under natural convection conditions is unclear and both enhancement and deterioration have been observed, which suggests that additional understanding is required. Suggestions for the observed behavior have been provided in terms of electrostatic behavior of the nanoparticles in the base fluid and possible sedimentation. However, there is no conclusive experimental evidence for all the suggested sources influencing nanoparticle behavior and heat transfer rate and this forms the purpose of the currently reported research. The remaining paper presents the experimental arrangement and methodology in the next section, the results in terms of heat transfer performance – absolute and normalized heat transfer coefficient, Nusselt and Rayleigh numbers under different boundary conditions and for various nanoparticle volume fractions – and associated discussion in Section 3. Finally, a summary of the main conclusions is presented.

2. Experimental facility and procedure

2.1. Experimental setup

The experimental setup is attempting to generate a classical Rayleigh–Benard system, in order to measure the heat transfer coefficient under natural convection. It comprises a cubic cell with a volumetric capacity of $1 \times 10^{-3} \text{ m}^3$ with optical access through the side walls. A schematic diagram of the apparatus is shown in Fig. 1. The natural convection rig consists of many components that all together ensure a satisfactory operation, undisturbed flow fields and minimum heat losses. The bottom plate consists of an aluminum heating pan A of thickness 20 mm, which houses 5 parallel cartridge heaters of 300 W power output each. The heating pan A provides heat power to an insulating pan B that is placed on top and under the aluminum heating plate C. This configuration ensures that heat transfer from the heating plate in the downward direction is minimized. The heating plate C, which is 30 mm thick, consists of 4 parallel cartridge heaters of 80 W power output each. Around the heating plate, there is a Teflon plate D to eliminate any heat losses from the sides. The aluminum cubic cell E is the key component that houses the tested medium. It also provides perimetric optical access for possible visualization studies, by incorporating four quartz windows (2 large square [40 mm × 40mm] and 2 small rectangular [10 mm × 40 mm]). Around the cell, there are 4 specially designed insulating pans F, 3 mm thick, and also a transparent Plexiglas cover G of 10 mm thickness. This provides additional insulation and brings all the subcomponents of the chamber together. At the top of the test cell, there is an aluminum heat sink H connected with a cooling water supply. Finally, between the heat sink and the cubic cell, a Teflon plate I is placed to prevent their thermal connection.

Eleven thermocouples are placed at various locations inside and outside the chamber to monitor and evaluate heat transfer performance. From top to bottom, six thermocouples are placed at the heat exchanger. Two of them measure the temperature at the inlet and outlet of the cooling unit, whereas another four measure the temperature at the top, cold plate, 5 mm above the free surface of the cell. Two thermocouples are placed at the heating plate. The first one is vertically placed 6 mm below the cell's free surface to measure the temperature $T_0$ of the bottom, hot plate. The second is placed on the bottom of the plate and measures the temperature there. One thermocouple is attached at the top surface of the heating pan and is used as a key indicator of the heat flux provided by the pan in order to match the temperature of the heating plate and thus eliminate the heat losses downwards. Finally, two thermocouples are embedded inside the heating plate and heating pan and measure the temperature there, so as to prevent any overheating or failure of the cartridge heaters.

2.2. Control system

The operation of the nanofluid chamber was controlled through LabVIEW software, coupled with National Instruments (NI) hardware and supplementary in-house electrical device. Through the
software, the operation of the rig was monitored and controlled in real time, while the data, namely input power, energy losses and temperatures, were stored at a sampling rate of 1 Hz.

The NI hardware consists of a CompactDAQ chassis (cDAQ-9188), which integrates connectivity and signal conditioning into modular I/O for direct interfacing to any sensor or signal, and voltage input (NI 9213) and output (NI 9263) modules. The additional in-house electrical device, which is connected to the CompactDAQ chassis and the natural convection chamber, consists of two power controlling thyristors (United Automation PR1-DIN-2.5KW) and two temperature controllers (TC Direct 309100). Each thyristor corresponds to a heater assembly – one for the heating plate and the second for the heating pan. Both thyristors are connected to the NI voltage output module and the heaters (heating plate and pan). The temperature controllers are connected with two K-type thermocouples, located inside the heaters. Through an improved autotuning algorithm, they achieve fast and stable temperature control and prevent possible damages to the heaters and the rig in the unexpected case of over-heating.

The LabVIEW software incorporates different operating scenarios according to the selected boundary conditions. In this study, constant heat flux at the heating plate and constant temperature at the cooling unit were enabled. The main feature in this operating scenario is a virtual proportional–integrate–derivative (PID) controller that matches the temperature of the heating pan to the heating plate, hence eliminating any heat losses in the downward direction.

2.3. Nanofluid preparation

In this study, no pH changers or surface activators were added during any stage of the nanoparticle production or the synthesis of the nanofluid as these could affect the thermophysical properties of the suspension [20]. For instance, Eastman et al. [32] reported that in samples where a small amount of thioglycolic acid (<1 vol.%), were synthesized and tested for all three power inputs. The heat transfer results of this study were considered and compared under varying induced flow regimes. The minimum power input was set at 150 W, which ensures that the maximum heat losses from the chamber are less than 5% of the input heat flux. The intermediate power input was set at 150 W and the maximum at 175 W, since beyond this power boiling is initiated. γ-Al2O3–DI H2O nanofluids of various concentrations, ranging from 0.01 to 0.12 vol.%, were synthesized and tested for all three power inputs. The heat transfer results of this study were considered and compared under steady state conditions. In the absence of pH changers and dispersants, there was a need to perform the nanofluid experiment fast enough to eliminate any issues related to the stability of the suspension. For this reason, the nanofluid experiments were performed when steady state conditions for pure DI water had been established.

A two-step preparation method was followed, according to which the nanoparticles are dispersed as a dry powder into the base fluid. The γ-Al2O3 nanoparticles had a nominal particle size of less than 50 nm (quantified by TEM), a particle density of 4000 kg/m³ and were supplied by Sigma Aldrich. Additional TEM measurements were performed in-house to identify the average particle size, shape and purity of the aluminum oxide nanopowder. For these measurements, a JEOL 2100F TEM operating at 200 kV was employed, according to the process described in Barrett et al. [6]. Fig. 2 presents TEM images for the aforementioned aluminum oxide powder. It can be observed that the nanoparticles are highly agglomerated and form large clusters. This was expected since no ultrasonication was performed and the nanopowder is initially in agglomerated form due to the strong Van der Waals forces. Also, this figure depicts both the size and shape of the nanoparticles. It can be seen that the identified ultra-fine particles look like nanoclay flakes and spheres with an average diameter of 10 nm. In addition, the material composition was measured and verified through TEM. In Fig. 3, spectral data for the nanopowder indicate a highly pure (~99%) aluminum oxide powder.

High purity DI water, supplied by VWR, was used as the carrier fluid. At the beginning of every experiment, the required amount of nanopowder was measured by weight, using a high accuracy electronic balance (Sartorius semi-microbalance R 200 D). Then, the powder was added to a 15 ml glass test tube and filled up with DI water. The next step was the ultrasonication process that is followed in order to break the agglomerates into their primary particle size. The reason is that the commercially available dry nanoparticles are initially in an agglomerated form due to the strong Van der Waals attractive forces among the particles [28,30,33]. An ultrasonic bath (Pulsatron KC2 by Guyson International Ltd) was used in combination with an analogue vortex mixer (mini vortex mixer by VWR) for a period of 5.0 h [6,29] to effectively break and disperse the weakly-bonded agglomerates by high shearing.

2.4. Methodology

The first step of the current study was a careful evaluation and characterization of the apparatus used, in order to select appropriate boundary conditions for the needs of this work. Three different power inputs at the heating plate were used to test the nanofluid under varying induced flow regimes. The minimum power input was set at 125 W, which ensures that the maximum heat losses from the chamber are less than 5% of the input heat flux. The intermediate power input was set at 150 W and the maximum at 175 W, since beyond this power boiling is initiated. γ-Al2O3–DI H2O nanofluids of various concentrations, ranging from 0.01 to 0.12 vol.%, were synthesized and tested for all three power inputs. The heat transfer results of this study were considered and compared under steady state conditions. In the absence of pH changers and dispersants, there was a need to perform the nanofluid experiment fast enough to eliminate any issues related to the stability of the suspension. For this reason, the nanofluid experiments were performed when steady state conditions for pure DI water had been established.

![TEM images of γ-Al2O3 nanoparticles; (a) large particle agglomerate and (b) average particle size and shape in the sample.](image-url)
been reached. The experimental procedure involved the deactivation of the heating elements for a short period of time (~5 min) and the removal of the top, cold plate. Then, the sonicated nanofluid samples were poured into the chamber and stirred well with the rest of the base fluid to ensure good dispersion. Finally, the heaters were switched on again and the rig was operated until steady state conditions were reached. At steady state (about 3.0 h after switching on the heaters), the equivalent temperatures of the heating and cooling plates were calculated by averaging a sample record of 1000 sets of temperature measurements recorded at a sampling frequency of 1 Hz via the corresponding hot and cold plate thermocouples. This averaging process is followed in order to eliminate the temporal and spatial temperature fluctuations on the plates arising due to turbulence and buoyancy effects. Subsequently, the heat transfer properties, namely heat transfer coefficient, Nusselt and Rayleigh numbers were calculated.

3. Results and discussion

In this part, results of the heat transfer performance under natural convection of nanofluid are presented and compared with DI water. More specifically, the absolute and normalized (with the value for DI water) heat transfer coefficient as a function of nanoparticle volume fraction and power input is depicted. In addition, Rayleigh number in terms of power input and Nusselt as a function of Rayleigh number are illustrated to identify their dependency and evaluate the resulting heat transfer performance.

The section is divided into six subsections; at first, the quality of the nanofluid is evaluated in terms of stability, by performing a qualitative visualization study of a ‘cold’ nanofluid placed in a glass flask. Following, the thermophysical properties of the test fluids are considered by including some widely available correlations used in the present study. Then, the temporal evolution of the temperature difference between the heating and cooling plate until steady state conditions is presented for both water and nanofluid. Finally, the influence of the nanoparticle volume fraction as well as the heat flux on the heat transfer properties of the nanofluid is investigated and finally, an extensive discussion on the findings is included.

3.1. Nanofluid stability

For the study of the thermophysical properties of nanofluids, pure nanosuspensions were used without any surface additives or pH controllers. Thus, any mechanism responsible for the altered thermophysical properties of nanofluids can be better identified and studied. Therefore, in such cases, the purity of DI water as well as its initial pH value play dominant roles. The quality of water is strongly associated with the followed purification process. Therefore, technical standards on water quality have been established by many national/international organizations. One such is the international organization for standardization (ISO), according to which there are certain limits to categorize water in terms of purity. In this study, high purity DI water (grade 3 according to ISO 3696 standard) that is produced by distillation and capacitive deionisation was used with an initial pH value of 6.9. However, both the introduction of Al2O3 nanoparticles in the base fluid and the CO2 of the atmosphere affect the final pH value of the nanofluid.

A visualization study, Fig. 4, was conducted to qualitatively assess the stability of a freshly prepared nanofluid and record any possible sedimentation or separation in terms of time. The study was performed for a ‘cold’ nanofluid (no heat flux was induced), placed in a 1.0 L rectangular glass flask that approaches the shape of the chamber’s main body. It can be observed that after 24 h, no visible change in the nanofluid stability took place. However, in the middle of the first week after the preparation of the nanofluid mixture, the suspension became less bright, indicating significant nanoparticle deposition. During the period between the first half week and up to the second week, the nanofluid appeared qualitatively unchanged in terms of stability, while in the third week concentration stratification layers were visible. In addition, deposition of aluminum nanoparticles or agglomerates was noticed at the bottom of the flask even during the first few hours.

3.2. Thermophysical properties of nanofluids

The Al2O3–H2O nanofluid is a well-studied suspension concerning its physical properties. The water properties, namely dynamic viscosity, \( \mu_w \), the thermal conductivity, \( k_w \), the heat capacity, \( c_p \) and the thermal expansion coefficient, \( \beta \), were obtained from Rohsenow et al. [34]. As for the nanofluid, they were calculated by widely used empirical correlations and formulas from the literature. Also, both the properties of water and nanofluid were considered at the mean temperature of the hot and cold plates.

The density of a nanofluid, \( \rho_{nf} \), can be obtained from the following widely accepted formula [2,5,19,35–39] which relates the
density of the base fluid, \( \rho_w \) with the density and the volume fraction of the nanoparticles, \( \rho_n \) and \( \varphi \), respectively

\[
\rho_{nf} = (1 - \varphi)\rho_w + \varphi\rho_n
\]

In a similar way, the heat capacity of the nanosuspension, \( c_{p,nf} \), can be expressed by the extensively used correlation \[2,35,40\]:

\[
c_{p,nf} = \frac{\varphi c_{p,n} + (1 - \varphi)\rho_w c_{p,w}}{(1 - \varphi)\rho_w + \varphi\rho_n}
\]

For the nanofluid viscosity, \( \mu_{nf} \), we adopt an empirical formula that was initially obtained by Maiga et al. \[41\] but also verified and used by Ni et al. \[2\]

\[
\mu_{nf} = \mu_w[1 + 7.3\varphi + 123\varphi^2]
\]

It is based on experimental data using aluminum based nanofluid with an average particle diameter of 20 nm, which was very close to ours, for room temperature. Nevertheless, it was experimentally observed that the measured viscosity did not have a strong temperature dependency and thus gave good representation over a wide range of temperatures.

Finally, for the conductivity, \( k_{nf} \), a fitting formula used by Ni et al. \[2\], which is based on an empirical relationship \[41\], was involved. This has been evaluated with different \( \text{Al}_2\text{O}_3-\text{H}_2\text{O} \) nanofluid data sets over comparable ranges of temperature and concentration with the present work:

\[
k_{nf} = k_w[1 + (-49.796 + 0.1787T)\varphi + (535.576 - 1.840T)\varphi^2]
\]

In the current investigation the particle size involved was smaller than that of Ni et al. \[2\], but for relatively low nanoparticle concentrations – such as those used in this study – Eq. \(4\) can be approximately reduced to the theoretical correlation from the effective medium theory \[2,42\]. Therefore, no significant augmentation of the nanofluid thermal properties was expected.

In free convection applications, the heat transfer correlations are of the form of \( Nu = f(Gr, Pr) \), where the Nusselt number (\( Nu \)) is a function of Grashof (\( Gr \)) and Prandtl (\( Pr \)) numbers. The \( Nu \) number relates the convective with the conductive heat transfer mode and is given by

\[
Nu = \frac{hL}{k}
\]

On the other hand, \( Gr \) governs the transition in the free convection boundary layer and \( Pr \) controls the relative thickness of the momentum and thermal boundary layers. These are given by

\[
Gr = \frac{g\beta\Delta TL^3}{\nu^2}
\]

\[
Pr = \frac{\nu}{\kappa}
\]

The product of the \( Gr \) and \( Pr \) numbers is known as Rayleigh number, \( Ra \) and is commonly used for free convection applications. This is given by,

\[
Ra = \frac{g\beta\Delta TL^3}{\nu^2}
\]

At steady state conditions, it is assumed that the temperature at the heating and cooling plates is uniform due to the highly conductive material of the aluminum plates and the insulation Teflon plates that surround them. Thus, the heat diffusion equation was adopted to calculate the surface temperatures \( T_{hu} \) and \( T_{cl} \):

\[
T_{hu} = T_h - \frac{Qd_h}{k_{pl}}
\]

\[
T_{cl} = T_c + \frac{Qd_c}{k_{pl}}
\]

As a result, the heat transfer coefficient is given by,

\[
h = \frac{Q}{A(T_{hu} - T_{cl})}
\]

### 3.3 Transient and steady state of the chamber wall temperature

The experiments were conducted until steady state conditions were reached and temperature signals were acquired and recorded every second. Due to the methodology followed for the nanofluid, no direct comparison can be made concerning the time required to achieve steady state between pure water and nanofluid. Steady state condition is assumed to be attained when the temperature difference between the hot and the cold plates becomes constant.

![Fig. 4. Stability of Al₂O₃–water nanofluid (φ = 0.06 vol.%) as a function of time.](image)

![Fig. 5. Temporal evolution of the time-averaged temperature difference between the heating and cooling plate for power input of 125 (circles), 150 (rhombus) and 175 W (squares), indicating the transient behavior and the steady state conditions, for pure DI water.](image)
and the energy rate absorbed by the cooling unit reaches a constant value. In Fig. 5, for DI water, a nearly constant temperature difference is reached after an hour for all three input power inputs. However, steady state conditions are reached after 3.0 h where the power absorbed by the cooling stabilises at a particular level, as depicted on Fig. 6.

3.4. Influence of nanoparticle volume fraction on heat transfer properties

The natural convective heat transfer coefficient strongly depends on the geometry of the convective chamber as well as on the thermophysical properties of the testing fluids. Some additional factors are the configuration and orientation of the heating elements, the method of heating and the roughness of the chamber’s walls [30]. In this study, pure DI water and nanofluid experiments were conducted for three different boundary conditions and various nanoparticle volumetric concentrations, namely 0.01%, 0.03%, 0.06% and 0.12%. The main purpose was to investigate the convective heat transfer performance of nanofluids and to study the influence of the nanoparticle concentration in the suspension. As a first step, constant heat flux, \( q^0 = 19.5313 \text{ kW/m}^2 \) (corresponding to input power of 125 W) was applied to the bottom plate and constant temperature, \( T_c = 21^\circ \text{C} \) (average value) was imposed to the cold plate to study the heat transfer performance and the flow dynamics of the nanofluid as a function of the nanoparticle volumetric concentration. In Fig. 7, the normalized heat transfer coefficient of nanofluid to that of water for various nanoparticle concentrations is presented at constant power input of 125 W (circles). It can be observed in Fig. 7 that the addition of aluminum nanoparticles to the aqueous base fluid deteriorates the heat transfer coefficient. As the nanoparticle concentration increases, the heat transfer coefficient decreases. Also, even though the flow becomes more turbulent due to the addition of nanoparticles, the convective heat transfer mode is reduced. The next step involved the examination of the impact of more intense turbulent flows on the heat transfer performance of the nanofluid as the nanoparticle concentration was increased. For this case, heat flux, \( q^0 = 23.4375 \) and 27.3438 kW/m² (corresponding to input power of 125 and 175 W, respectively) at the bottom plate was imposed, while the temperature of the cold plate was maintained constant, \( T_c = 21^\circ \text{C} \) (average value). In Fig. 7, the normalized heat transfer coefficient of nanofluid to that of water for various nanoparticle concentrations is presented at constant power, \( Q = 150 \text{ W} \) (rhombus) and 175 W (squares). Even under a more intense turbulent flow, the addition of nanoparticles results in deterioration of the heat transfer performance relative to the base fluid. For all three power inputs, the non-dimensional heat transfer parameters are presented in Table 1.

3.5. Influence of the input heat flux on the thermophysical properties

In this section, water and nanofluid data are compared for the nanoparticle volumetric concentrations, \( \varphi = 0.03\% \), 0.06\% and 0.12\% for all power inputs, \( Q \), of 125, 150 and 175 W. The purpose is to identify any changes in the absolute value of the heat transfer coefficient trendline as the imposed heat flux increases for pure water and nanofluid. In Fig. 8, the heat transfer coefficient of pure DI water and nanofluid in terms of the power input level is presented. It can be seen that as the power input increases, the heat

| Table 1 |
|---|---|---|---|---|---|---|---|
| Oper. scenario | \( Q \) (W) | \( q^0 \) (kW/m²) | \( \varphi \) (vol.%): | \( \Delta T \) (°C) | \( h \) (W/m²K) | \( Nu \) | \( Pr \) | \( Ra \times 10^3 \) |
| A | 125 | 19.5313 | 0.00 | 52.53 | 372 | 58.0 | 3.7 | 2.5 |
| | | | 0.01 | 53.70 | 364 | 56.9 | 3.7 | 2.6 |
| | | | 0.03 | 54.15 | 361 | 56.3 | 3.7 | 2.6 |
| | | | 0.06 | 55.26 | 353 | 54.9 | 3.7 | 2.7 |
| | | | 0.12 | 57.05 | 342 | 53.3 | 3.7 | 2.7 |
| B | 150 | 23.4375 | 0.00 | 61.63 | 380 | 59.1 | 3.4 | 3.6 |
| | | | 0.01 | 63.77 | 368 | 57.0 | 3.4 | 3.6 |
| | | | 0.03 | 63.05 | 372 | 57.5 | 3.4 | 3.5 |
| | | | 0.06 | 63.31 | 370 | 57.1 | 3.4 | 3.5 |
| | | | 0.12 | 66.70 | 351 | 53.7 | 3.1 | 4.1 |
| C | 175 | 27.3438 | 0.00 | 67.74 | 404 | 62.4 | 3.1 | 4.2 |
| | | | 0.01 | 68.50 | 399 | 61.3 | 3.0 | 4.6 |
| | | | 0.03 | 68.27 | 401 | 61.5 | 3.0 | 4.5 |
| | | | 0.06 | 70.79 | 386 | 59.0 | 3.0 | 4.7 |
| | | | 0.12 | 77.54 | 353 | 53.5 | 3.0 | 5.2 |
transfer coefficient increases for all the cases. However, for the maximum nanoparticle concentration the rate of enhancement is notably less.

In Fig. 9, the Rayleigh number, $Ra$, is presented as a function of the power input level for water and nanofluid. As the nanoparticle concentration and the power input level are raised, the flow becomes more and more turbulent, as revealed by the increase of the $Ra$. Nonetheless, the increment is near 20% for the maximum nanoparticle concentration. In Fig. 10, $Nu$ for water and nanofluid as a function of $Ra$ is depicted. In most of the cases, as $Ra$ increases, $Nu$ increases. However, for the maximum nanoparticle concentration, $Nu$ becomes nearly constant. The observed $Ra$–$Nu$ dependency indicates a relationship of the type,

$$Nu = cRa^n$$

(12)

which has also been presented by other numerical and experimental studies [2,43,44]. In this work, the constant “$c$” and the index “$n$” appear to be strongly related to the nanoparticle concentration and the stability characteristics of the suspension. For the nanofluid with maximum nanoparticle concentration, the relationship of Eq. (12) is no longer valid, due to the significant nanoparticle deposition on the hot, bottom plate.

3.6. Repeatability test and error analysis

To ensure reliability of the averaged results for DI water, the uncertainty in the mean value of the heat transfer coefficient, $\sigma_w$ was calculated according to Kirkup [45] and found to be 2 W/m$^2$ K for all three power inputs. For the nanofluid, due to time and cost limitations, it was not possible to perform such an extended analysis for every nanoparticle volume fraction, for all three power inputs. Therefore, a repeatability test was performed for an arbitrary selected nanofluid of a specific nanoparticle volume fraction. The purpose was to ensure repeatability of the nanofluid results and to identify whether any systematic uncertainties are induced either by the nanofluid preparation or the experimental process followed. For the 0.03 vol.% nanofluid under 125 W, a set of five individual measurements was considered and subsequently, a comparison of the temporal evolution of the temperature difference between the heating and cooling plate for the whole set was performed. As illustrated in Fig. 11, the temperature differences at steady state conditions are very close to each other, with the standard deviation being 0.59 °C. Concerning the uncertainty in the mean, $\sigma_{ref}$ for the heat transfer coefficient, it was found to be the same with that of DI water, 2 W/m$^2$ K. Through the repeatability tests, the uncertainty of the measured value of the heat transfer coefficient was found to be very small and as a consequence, the individual calculated values are considered highly precise.

The calculation of the uncertainty in both the values and numbers that characterize the thermal performance of liquids requires the determination of the uncertainties in the measured quantities through error propagation. Main sources of uncertainty in the calculations were the resolution of the flow meter employed at the cold plate, ±0.055 L/min used to calculate the heat losses from the sides of the rig, the tolerance of the thermocouples, ±1.1 °C of...
the reading value, and the resolution and measurement error of the control instrumentation, ±0.9 °C for the temperature range of interest. In this study, where simple arithmetic calculations were required, the mean uncertainty in the heat losses from the sides of the rig, for instance, were calculated by,

$$\Delta(P) = \Delta(F) = \frac{|\partial F|}{\partial W} \Delta(W) + \frac{|\partial F|}{\partial T} \Delta(T)$$  \hspace{1cm} (13)$$

where it was found to be 7.7 W. In a similar way, the mean and the maximum fractional uncertainty (%) of the heat transfer coefficient were 6.7% and 7.9% respectively.

3.7. Discussion

An important finding of this study is that the addition of γ-Al2O3 nanoparticles to pure DI water deteriorates the natural convective heat transfer performance under the specific experimental conditions. This result is consistent with other experimental observations in the literature, where the heat transfer coefficient, h and Nu decrease with the addition of nanoparticles, even though Ra increases. However, natural convective heat transfer deterioration has been reported not only for Al2O3–H2O nanofluids [2,9,21–22], but also for CuO [9] and TiO2–H2O nanofluids [20]. Such a finding is contradictory to the general expectation that the heat transfer in nanofluids increases. In the vast majority of nanofluid studies for conduction [29,46–58], forced convection [12,59,60], boiling [28,61] and critical heat flux [62–64], heat transfer enhancement was reported. Even more, heat transfer increase was observed in many numerical natural convection studies, making the overall behavior of nanofluids even more questionable and unclear. Up to date, the reported heat transfer deterioration under natural convective experimental studies is attributed to some widely addressed, but not systematically verified mechanisms, suggested in the literature. Savithiri et al. [65] performed a scaling analysis for the investigation of slip mechanisms in nanofluids and they concluded that properties like the shape, size, concentration and temperature of the nanoparticles have major effects on the slip forces. Some widely reported mechanisms, responsible for the heat transfer deterioration in natural convective studies, include: the particle–fluid slip, the particle–surface and particle–particle interactions, the convection induced by particle concentration differences, the reduction of Ra due to the increased kinematic viscosity, the Brownian motion, the thermophoretic motion of nanoparticles and the sedimentation. Some speculated proposed mechanisms, attempting to explain the alteration (enhancement and/or reduction) of the general heat transfer performance of nanofluids, can be found in the extended literature review by Sergis and Hardalupas [7]. However, there is not yet conclusive experimental evidence to support them.

In the present experimental investigation, the fouling of the bottom surface appears to play dominant role for the deterioration observed. This is mainly due to the inadequacy of the system forces to ensure both long and short-term nanofluid stability, in the absence of any surfactant or pH changer. Therefore, strong attractive Van der Waals forces between the particles present in the suspension result in the formation of nanoparticle agglomerates that could lead or accelerate nanoparticle settling at the bottom of the rig. Also, as the nanoparticle volumetric concentration increases, the temperature at the heating plate increases. This can be credited to the increased fouling of the heating surface due to the formation of a thin stationary layer structure near the bottom that consists of nanoparticles, agglomerates and water molecules and impose additional thermal resistance in the system that obstructs the efficient heat transfer. This observation could be qualitatively verified for the first time, since the experiments were performed within a chamber that had optical access. Fig. 12 illustrates severe nanoparticle deposition at the bottom of the chamber for a highly unstable nanofluid, γ-Al2O3 nanoparticles suspended in low-quality DI water, with a pH value that approaches the IEP of aluminum oxide. It is observed that a thick layer of nanoparticles and water molecules has been formed at the bottom of the chamber that appears to be responsible for the increased temperature at the heating plate. Fig. 13 provides images that demonstrate the layer formation within the chamber for a nanofluid of maximum nanoparticle concentration, 0.12 vol.%. More specifically, Fig. 13 (a) shows the deposition of nanoparticles at the bottom plate of the rig, verified when pumping the nanofluid out of the chamber, at the end of the experiment, where Fig. 13(b) depicts the fouling of the heating surface with nanoparticles and agglomerates that remain adhered to the surface of the heating plate. It was also noticed that when the imposed heat flux was increased, the adhesion of nanoparticles at the heating plate was more intensive. This can be mainly credited to the thermophysical properties of the γ-Al2O3 nanopowder used in this study and to the resulting interactions between the heating surface and the deposited nanoparticles and agglomerates at the bottom, caused by the reduction of surface’s wettability. To summarise, particle–particle and particle–fluid interactions that led to significant sedimentation as well as particle–surface interactions between the nanoparticles and the heating surface appear to be the major mechanisms that affected the heat transfer behavior under natural convection in the current investigation. In similar studies, such as the one conducted by Ni et al. [2], where a very small amount of dispersant was added in the suspension to minimise nanoparticle agglomeration the decrease of the heat transfer coefficient in nanofluids was not that
intense, even for higher nanoparticle concentrations. In their experimental investigation, sedimentation seems not to be a major deteriorating mechanism, as the $Ra$ number decreases with the increasing volume fraction of nanoparticle in the base fluid.

The effect of fouling of the heating surface on the heat transfer coefficient of nanofluid is modeled numerically, in order to predict the temperature gradient between the hot and cold plates. The significance of the fouling effect is evaluated qualitatively to verify the experimental results. To start with, a nanofluid of maximum nanoparticle concentration 0.12 vol.% is considered under the operating conditions A (Table 1). At steady state conditions, a thin film that consists entirely of $Al_2O_3$ is assumed to have uniformly covered the heating plate of the rig. Therefore, an additional thermal resistance should be also considered in the heat transfer calculations for nanofluid, according to Eq. (14).

$$Q = \frac{\Delta T_w}{k_A} = \frac{\Delta T_{layer}}{k_{layer}} = \frac{\Delta T_{nf}}{k_{layer} + \frac{\rho}{\rho_{nf}}} \tag{14}$$

By substituting the values from Table 1 and assuming a small layer thickness $L \approx 0.200 \text{ mm}$ and $k_A = 40 \text{ W/mK}$, it is found that $\Delta T_{layer} < 0.10 \text{ °C}$. Therefore, nanoparticle deposition on its own cannot explain the increased temperature of the heating surface and thus the reported convective heat transfer deterioration.

In a different approach for the fouling effect of the heating plate, a thin layer that contains nanoparticles, agglomerates and water molecules is considered. Like the previous analysis, nanofluid of maximum nanoparticle concentration, 0.12 vol.% is considered under the operating conditions B (Table 1) at steady state. Suppose that 25% of the nanoparticles, for instance, have settles out of the suspension and deposited close to the heating plate in a volume of $100 \times 100 \times 0.2 \text{ mm}^3$ that corresponds to a nanoparticle concentration of $\phi = 15 \text{ vol.\%}$ in that layer. Therefore, by substituting the values from Table 1 and taking into account that $L = 0.200 \text{ mm}$ and $k = 0.643 W/\text{mK}$ at $20 \text{ °C}$ and $k_{layer} = 0.93 W/\text{mK}$ (calculated from the effective medium theory, where $\frac{k}{k_{layer}} = 1 + 3\phi$), it is found that $\Delta T_{layer} = 4.20 \text{ °C}$ and $\Delta T_{nf} = 56.73 \text{ °C}$. By comparing this value with the experimentally measured, 57.05 °C, excellent agreement is noticed. By following the same procedure, but under the operating conditions B (Table 1), it is reported that $\Delta T_{layer} = 5.04 \text{ °C}$ and $\Delta T_{nf} = 66.67 \text{ °C}$, which is very close to the experimentally measured, 66.70 °C. Finally, the same process is performed for operating conditions C (Table 1), where $\Delta T_{layer} = 5.88 \text{ °C}$ and $\Delta T_{nf} = 73.62 \text{ °C}$, while the experimentally observed is 77.54 °C.

Therefore, the experimentally reported effects of fouling near the heating plate at the bottom of the rig can be modeled successfully, regardless of the assumptions considered for the thickness or the concentration of that layer. As a consequence, in stationary liquids and low-velocity applications, special attention should be paid to the nanoparticle layer structure that results in confined liquid contact with the surface and thus limited surface wettability. Therefore, when low-velocity applications, such as natural convection ones, are involved special attention should be paid on ways to prevent or eliminate the formation of such layer structure. This can be done primarily by improving the attained stability of the suspension, through the employment of stabilization methods, such as the electrostatic. In this way, the surface charge of the nanoparticles can be increased, strong repulsive forces among them will be established and, as a consequence, any possible agglomeration will be eliminated. To summarise, a combination of a proper selection of nanopowder, in terms of its physical characteristics, along with the addition of a pH changer could provide a first solution towards this direction.

Acknowledgements

This work was funded by the RCUK Energy Program and EURATOM. The views and opinions expressed herein do not necessarily reflect those of the European Commission.

We would like to thank CCFE for the financial support through the CASE EPSRC studentship that allowed this research and especially Dr. Barrett Thomas for his input to this project. Finally, we would like to thank Dr. Byron Chan for his valuable help in designing and manufacturing the thermal rig. Data supporting this publication can be obtained by request from MultiphaseFlows@imperial.ac.uk.

References

[1] J. Milone, A. Burns, D. Drikakis, Computational modelling of the HyperVapotron on cooling technique, Fusion Eng. Des. 87 (9) (2012) 1647–1661.
[2] R. Ni, S.-Q. Zhou, K.-Q. Xia, An experimental investigation of turbulent thermal convection in water-based alumina nanofluid, Phys. Fluids 23 (2) (2011) 022005.
[3] S.U.S. Choi, J.A. Eastman, Enhancing thermal conductivity of fluids with nanoparticles, Dev. Appl. Non Newtonian Flows 66 (99–105) (1995).
[4] S.U.S. Choi, Nanofluids: from vision to reality through research, J. Heat Transfer 131 (3) (2009) 1–5.
[5] J. Buongiorno, Convective transport in nanofluids, J. Heat Transfer 128 (3) (2006) 240–250.
[6] T.R. Barrett, S. Robinson, K. Flanders, A. Sergis, Y. Hardalupas, Investigating the use of nanofluids to improve high heat flux cooling systems, Fusion Eng. Des. 88 (9–10) (2013) 2594–2597.
[7] A. Sergis, Y. Hardalupas, Anomalous heat transfer modes of nanofluids: a review based on statistical analysis, Nanoscale Res. Lett. 6 (1) (2011) 391.
[8] D. Wen, C. Lin, S. Vafaei, K. Zhang, Review of nanofluids for heat transfer applications, Particuology 7 (2) (2009) 141–150.
[9] N. Prata, W. Roetzel, S.K. Das, Natural convection of nano-fluids, Heat Mass Transfer 39 (2003) 775–784.
