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ORIGINAL

Experimental characterization of convective heat transfer with MWCNT based nanofluids under laminar flow conditions

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Abstract This study describes an investigation on the convective heat transfer performance of aqueous suspensions of multiwalled carbon nanotubes. The results suggested an increase on heat transfer coefficient of 47 % for 0.5 % volume fraction. Moreover, the enhancement observed during thermal conductivity assessment, cannot fully explain the heat transfer intensification. This could be associated to the random movements among the particles through a fluid, caused by the impact of the base fluid molecules.

1 Introduction

In most industrial applications, an efficient transfer of heat is of utmost importance. Frequently, it is chosen a fluid as a medium for the heat transfer, being convection the most relevant heat transfer mechanism. Regarding the Newton's law of cooling, the pursuit of the thermal engineers lies upon the maximization of the heat transfer rate for a given temperature difference. However, the heat transfer coefficient is a complex function of the surface geometry, fluid flow rate and respective thermo-physical properties. The latter is influenced by the thermal conductivity in a most direct way, as this determines the overall thermal transport.

Conventional fluids, such as water, engine oil and ethylene glycol are commonly used as heat transfer fluids. Although various techniques have been developed over the years to improve heat transfer capabilities of the conventional fluids, their low thermal conductivity diminishes the performance enhancement and the high compactness of heat exchangers for a large number of applications [1]. It is well known that the thermal properties of conventional fluids can be enhanced through the suspension of millimetre or micrometre sized solid particles. However, its usage has not been reported as heat transfer fluids due to its fast sedimentations, erosion, fouling and increasing pressure drop for pumping the fluid [1–4].

In 1993, Grimm obtained a patent related to the thermal conductivity improvement of a fluid containing dispersed solid particles of Al measuring 80 nm to 1 μm. Despite the 100 % thermal conductivity enhancement claimed, the suspension presented a rapid settling [5]. Two years later, Choi et al. [6] reported that an innovative class of heat transfer fluids could be engineered by suspending metallic nanoparticles in conventional fluids, appearing for the first time the word nanofluid.

Therefore, a nanofluid is a suspension of ultra-fine particles (nanometre scale) in a conventional base fluid that anomalously enhances the heat transfer characteristics of the original base fluid [7, 8]. They are expected to be ideally suited in practical applications as their use incurs little or no penalty in the pressure drop due to the ultra-fine sizes' distribution of the nanoparticles. Moreover, these appear to behave as single-phase fluid than a solid–liquid

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mixture [9, 10]. Therefore, they are considered as an innovative class of heat transfer fluids, as they offer new possibilities to enhance heat transfer performance of energy systems.

The inclusion of different types of metallic and oxide nanoparticles on base fluids has been investigated by several researchers [6–8, 11–14]. For instance, Liu et al. [14] reported an increase in thermal conductivity of 60 % for nanofluids consisting of water and 5 % volume fraction of CuO nanoparticles. Murshed et al. [15], reported an enhancement of 33 % on thermal conductivity for 5 % volumetric loading of TiO_2 nanoparticles in water. Nevertheless, from experimental results, multiwalled carbon nanotubes (MWCNT) appear to be those that higher enhancement produces on the thermal conductivity of the nanofluids, for the same nanoparticles concentration [16]. This is a fundamental feature to avoid an undesired increase of the fluid viscosity and pressure drop, observed at higher nanoparticles volume fractions [17, 18].

Over the years, a broad number of researchers proposed several mechanisms in an effort to explain the thermal conductivity enhancements observed in suspensions. Bruggeman studied the thermal conductivity of micro and macroscale suspensions and found that the increased interparticle interaction contributes directly for the enhancement of the effective thermal conductivity [19]. These interactions are related to the spatial distribution of the particles and for high aspect ratio suspensions lead to the formation of percolation structures [20–22]. However, the nature in how these structures are formed is still an uncertainty among researchers [23]. The Brownian motion is suggested by many authors was the key mechanism governing the thermal behaviour of nanoparticles suspended in a base fluid [24, 25]. The latter, explains the random movement among the particles through a fluid, cause by the impact of the base fluid molecules. Lamas et al. [21], studied the effect of size, shape, aspect ratio and the Brownian motion of the nanoparticles to the interparticle interaction of CNTs suspension in a base fluid. They conclude that the size, shape and aspect ratio of the nanoparticles are main factors to the formation of dynamic networks and the influence of the Brownian motion seems to be negligible to the results.

Though, the development of nanofluids still encounters several obstacles such as the lack of agreement between results, poor experimental characterization of suspensions and the lack of theoretical understanding of the mechanisms responsible for the observed results [26]. Moreover, a full understating of their convective heat transfer features is also essential, despite a higher attention given by the research community to the thermal conductivity [27].

As known, the convective heat transfer coefficient depends on thermal conductivity specific heat capacity, viscosity, flow rate and density of the working fluids [28]. The fluid flow mode is of utmost importance, and most of the reported investigations characterize the heat transfer with emphasis on the convective heat transfer coefficient (h) or the Nusselt number Nu that directly depends on the Reynolds number (Re) and the Prandtl number (Pr). Some empirical attempts to compute the (Nu) as a function of Re and Pr numbers were performed both for laminar and turbulent flow regimes [29]. Several published experimental studies [10, 30–35], suggest that the Nu rises with the increase of particles volume fraction and Reynolds number. Several researchers [36–38] studied the convective heat transfer mechanism of nanofluids under laminar flow conditions, in horizontal tube heat exchangers, reporting a straight relationship between the enhancement of heat transfer and the aspect ratio of the MWCNTs.

In the last decade, MWCNTs have attained a great interest from the researchers community, due to their exceptional physical, thermal and electrical properties [39]. However, a major drawback of using MWCNTs lies with their poor dispersibility and homogeneity into the base fluid [40]. Nevertheless, recently Lamas et al. [41] reported a study on colloidal stability of nanofluids with covalently functionalized MWCNTs suspended in a water and ethylene glycol mixture, suggesting that those are stable for more than 50 years at rest conditions.

This paper presents original fundamental research work focus on the experimental characterization of the thermophysical properties of a family of MWCNTs based nanofluids namely convection heat transfer enhancement, under laminar flow condition, contributing to a better understanding of this new materials potential. The objective of the associated research line is the development of advanced heat transfer fluids, with significantly high thermal conductivity and convection coefficients without compromising other key properties, such as viscosity.

2 Experimental methodology

2.1 Materials

The studied MWCNTs were purchased to Cheaptubes Inc., and produced through catalysed chemical vapour deposition (CCVD). These have an outer diameter distribution ranging from 50 to 80 nm and a length distribution ranging from 10 to 20 µm, with a bulk density of 2.16 g/cm^3. Moreover, the selected base fluid was distilled water (DW). This choice is consistent with it importance for industrial applications as common heat transport fluid. Furthermore, the DW presents a satisfactory degree of purity, when compared to common water, allowing the reduction of noise factors.

2.2 Nanofluids preparation and colloidal stability

The production of a stable dispersion of MWCNTs is not a simple issue and requires the application of specific methodologies. At the nanoscale, the percentage of atoms at the particle surface becomes significant, increasing their surface-to-volume ratio. This characteristic induces strong attraction forces between the nanoparticles leading to the formation of local clusters [42, 43]. These clusters of increased size tend to settle down rapidly, leading to the degradation of the thermo-physical properties of the mixture. Therefore, MWCNTs require a previous covalent functionalization, as described by Esumi et al. [44]. The pristine MWCNTs were refluxed at 413 K in nitric and sulphuric acid at 1:3 volume ratio for 30 min, followed by washing with DW until no signs of acidity were found and dried in an oven at 373 K, for at least 72 h, to remove excess water. These functionalization places carboxylic groups on the sidewalls of the MWCNTs, which increases the repulsion forces of van der Waals, preventing the agglomeration [17, 41, 45]. Moreover, the carboxylic groups behave as weak acids and possess ion-exchange properties, having both hydrogen acceptors and hydrogen donors [46]. This will improve the MWCNTs wettability in aqueous solutions, due to a more hydrophilic surface structure.

In Fig. 1, is shown two scanning electron microscopy (SEM) images of (a) pristine and (b) covalently functionalized MWCNTs. As it can be depicted, the pristine MWCNTs (a) are highly agglomerated and, the covalently functionalized MWCNTs (b), are well dispersed moreover, the integrity of their tubular structure seems to be maintained.

The dried functionalized MWCNTs are dispersed in the base fluid through ultrasonication (Sonics and Materials 750 W ultrasonic processors @ 20 kHz) combined with a magnetic stirrer. The mixing time was previously determined to be 60 min [17, 45]. After these 60 min it was achieved a homogeneous distribution of the MWCNTs into the base fluid. Furthermore, nanofluids preparation methodology is strictly equal to that present by Lamas et al. [41]. MWCNTs based nanofluids with 0.25 and 0.5 % volume fractions were prepared through this methodology.

Colloidal stability of the prepared nanofluids was evaluated through UV–visible spectrophotometry (Shimadzu UV-mini 1240). The absorption measurement is based on the Beer–Lambert law [47–49], which is a linear relationship between the absorbance and the concentration of an absorber, and can be expressed as:

$$A = \varepsilon c l \tag{1}$$

where A is the measured absorbance, ε is the wavelength-dependent molar absorptivity coefficient, c is the concentration of the absorbing species in the solution and l is the path length. Moreover, through the mathematical formulation (2) it is possible to express the relative concentration of the samples with time [17].

$$\frac{A_t}{A_0} = \frac{\varepsilon c_t l}{\varepsilon c_0 l} = \frac{c_t}{c_0} \tag{2}$$

where the indices t and 0 represents, respectively, the measurement time step and the initial or zero time step.

The selected wavelength range was 200–500 nm and the maximum of absorbance for the MWCNTs was observed within 260 and 270 nm. In Fig. 2 it is shown the relative concentration variation over time for the 0.5 % volume fraction nanofluid. This suggests a convergence to 80 % of the initial concentration, achieved after 240 h, meaning that the nanofluid is stable after 24 h with a concentration decreasing of 20 %.

2.3 Thermo-physical properties assessment

The nanofluid effective thermal conductivity was measured through coated-transient hot wire (THW), which is the most widely used by the nanofluids' research community.

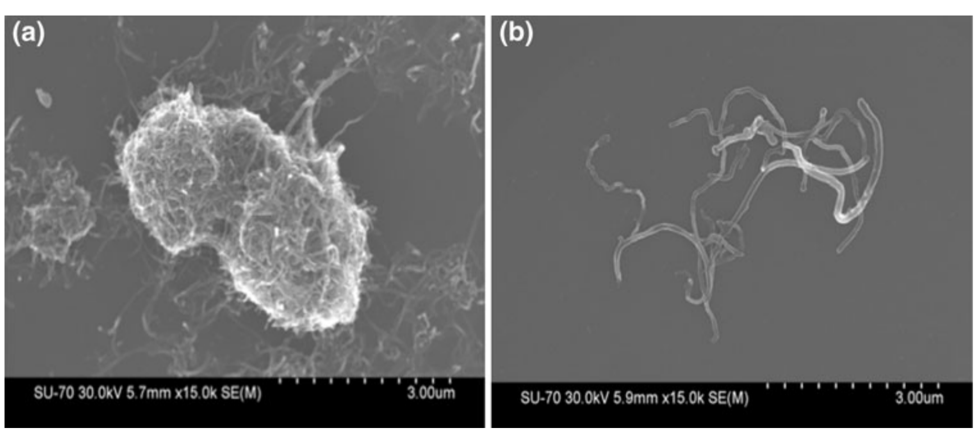

Fig. 1 SEM Pictures, of the **a** pristine MWCNTs and **b** covalently functionalized MWCNTs

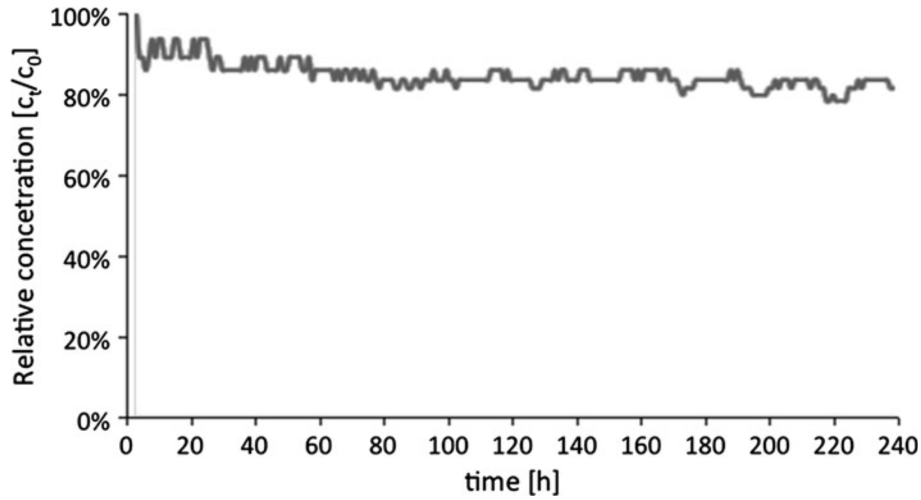

Fig. 2 Graphical representation of the relative concentration of the prepare nanofluid (0.5 % vol MWCNT/DW)

For that, a KD2 Pro thermal analyser (Decagon devices) was used. In order to increase the thermal stability of the measurements with temperature, a circulating liquid bath (with a Kryo30 solution) was attached to the measurement container. At least, 20 measurements were performed for each temperature ranging from 283.13 to 333.15, and for each nanofluid. To avoid external vibrations, the nanofluid sample was inserted in a double jacket container connected to the circulating bath and resting in a Styrofoam Box. In Fig. 3 it can be depicted the observed experimental thermal conductivity k variation with temperature for the base fluid (DW) and for the studied nanofluids. As it would be expected the thermal conductivity increases with particle volume fraction as well as with temperature. The obtained results suggest an average enhancement on the thermal conductivity of about 3 and 6 %, respectively.

The nanofluid viscosity was measured using an oscillation rheometer, for a shear rate ranging from 0 to 600 s^{-1} at 318.15 K. The considered viscosity for the calculation were 1.35×10^{-3} and 1.38×10^{-3} N s/m^2, for the 0.25 % vol MWCNT/DW and 0.5 % vol MWCNT/DW, respectively [45, 50].

The specific density (ρ) and heat capacity (Cp) where predicted through the analytical models suggested by Pak and Cho [29]. These can be expressed as:

$$\rho_{nf} = \varphi \rho_p + (1 - \varphi)\rho_{bf} \quad (3)$$

$$Cp_{nf} = \varphi Cp_p + (1 - \varphi)Cp_{bf} \quad (4)$$

where the indices *nf*, *p*, and *bf* are nanofluid, particle, and base fluid, respectively.

For the evaluation of the convection heat transfer coefficient, it was designed and constructed a working bench as shown in Fig. 4. The system is mainly composed by a reservoir, a peristaltic pump, a test section, a refrigerating zone, and a data-acquisition system for monitoring temperature. The test section is composed by a straight stainless steel circular tube with an inner diameter, D, of 6 mm and a length, L, of 1,200 mm. To ensure a constant heat flux, the heating system of the testing section is

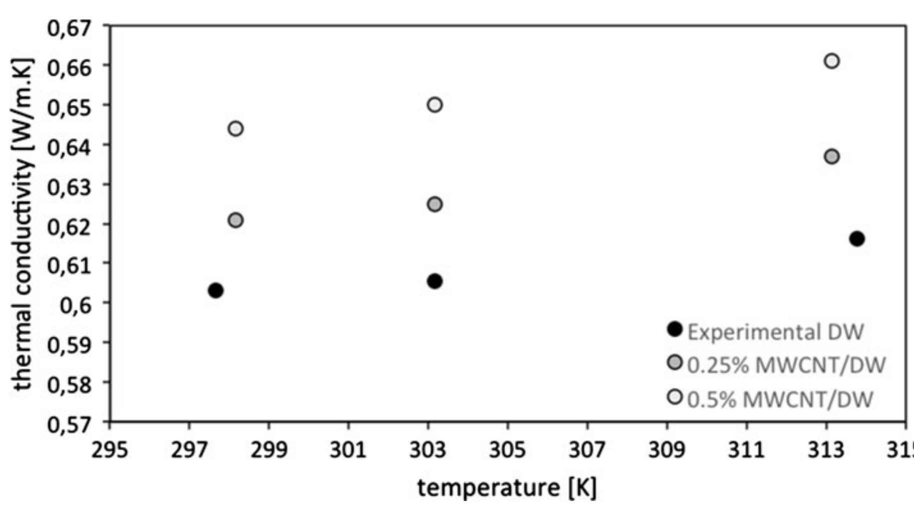

Fig. 3 Variation of experimental thermal conductivity as a function of temperature and particle volume fraction

composed by a silicone rubber heating tape of 432 W, rolled around the tube. To minimize heat loss from the heating tape to the environment, the test section was insulated by an aluminium tape and a tube of foamed polyethylene. Seven thermocouples were placed in the testing section to monitor both the fluid and the wall temperatures. Five of them attached on the external surface of the tube, to avoid flow interference, the remaining two immersed in fluid at the inlet and outlet sections of the testing tube. Prior to the test section, a hydrodynamic entry zone was dimensioned $(x_{fd,lam}) = 0.05\, Re_D D$ to accomplish a hydrodynamic fully developed flow at the entrance of the test zone. The refrigerating zone is composed by a tubular heat exchanger connected to a thermal bath. Two thermocouples are used to monitor the inlet and outlet fluid temperatures. This zone is also thermally insulated by a polyethylene foamed tube.

2.4 Convective heat transfer calculation

The total heat transfer to the system $(\overline{q''})$ is given by the following equation,

$$\overline{q''} = \frac{q'}{A_{sup}} \quad (5)$$

where q' is the heat energy provided to the system, and $A_{sup} = \pi DL$, is the heat transfer surface area.

Therefore, the experimental convection heat transfer coefficient is given by,

$$\overline{h_{exp}} = \frac{\overline{q''}}{T_s(x) - T_m(x)} \quad (6)$$

where $T_s(x)$, is the surface temperature on the axial point x and $T_m(x)$ is the average temperature of the fluid on the axial point x. The latter is given by,

$$T_m(x) = T_{in} + \frac{\overline{q''}\pi DL(x)}{\dot{m}Cp} \quad (7)$$

where T_{in} is the inlet temperature measurement, and $\dot{m}$ is the flow rate.

The Nusselt number can be described through the following equation,

$$Nu_{exp} = \frac{\overline{h}_{exp}(x)D}{k} \quad (8)$$

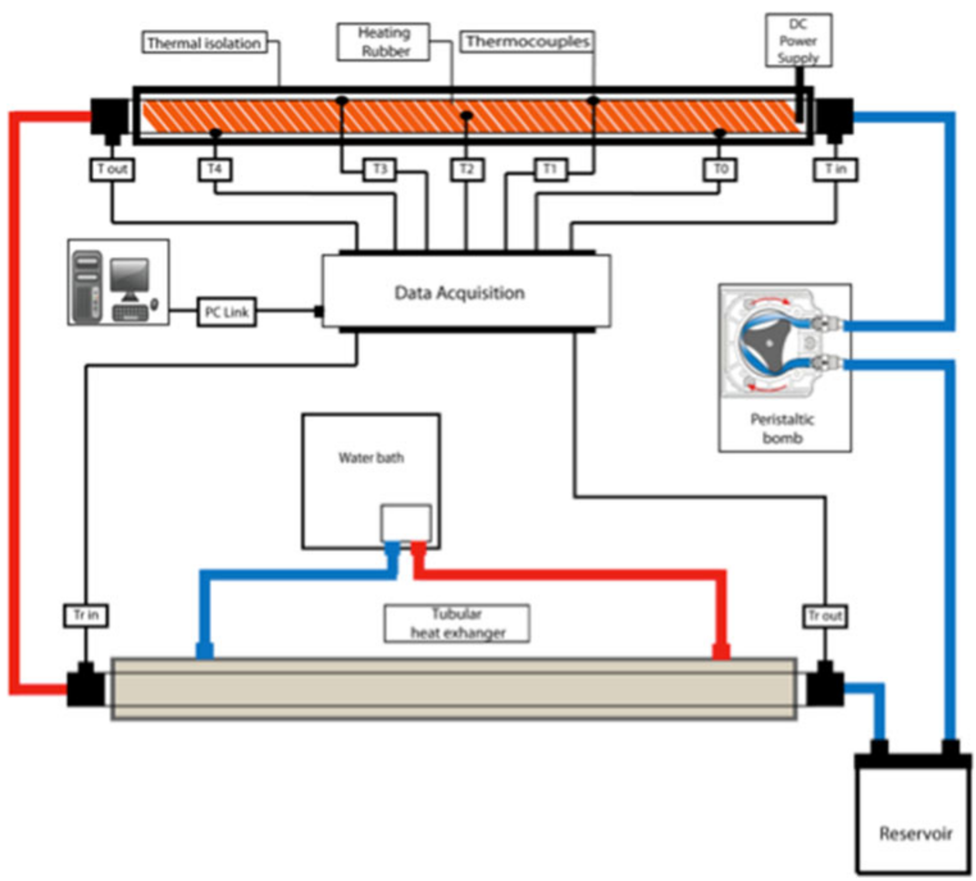

Fig. 4 Experimental setup of the heat transfer system for convection measurements

where k is the thermal conductivity of the fluid under measurement.

The Reynolds and Prandtl numbers are, respectively, given by [28],

$$Re = \frac{4\dot{m}}{\pi D \mu} \tag{9}$$

$$Pr = \frac{Cp\mu}{k} \tag{10}$$

where μ is the fluid under measurement viscosity.

In Table 1 are summarized the thermo-physical properties at 300 K.

The distilled water (DW) was the operating fluid chosen for the calibration process. This choice relies upon the fact that it is a fluid with well-known thermal properties. Three calibration tests were carried out on the system, in order to increase the confidence level on the overall experimental system:

1. Stability verification of the system through turning off the heating tape. A deviation less than 2 % between each thermocouple was observed.
2. Study of energy balance of the system. With heating tape powered at 108 W, and assuming that the external surface of the tube is adiabatic, the energy provided to the system presented a maximum deviation of 5 %.
3. The experimental apparatus was calibrated considering Shah equation for a laminar flow under a constant heat flux [51] and, a steady state, one-dimensional heat conduction model to couple the inner wall temperature and the measured temperature, Fig. 5. Accordingly to the proposed model, the temperature difference between the tube inner wall and the externally measure temperature is a function of the local heat flux and thermal resistances R_t, namely R_{t1} the contact resistance between the thermocouple and the tube external surface and R_{t2} the conduction resistance across the tube wall, Eq. 11.

$$T_{wall} = T_{measured} - q' R_t \tag{11}$$

where T_{wall} is the inner wall temperature, q' the local heat flux and R_t the total thermal resistance between the thermocouple actual location and the inner wall surface

Table 1 Thermal physical properties of the fluids at 300 K

Properties	Fluids		
	DW	0.25 % vol	0.5 % vol
ρ (kg/m^3)	997	1,197	1,398
Cp (J/kgK)	4,179	4,384	4,590
μ (Pa s)	8.55×10^{-4}	1.35×10^{-3}	1.38×10^{-3}
k (W/mK)	605×10^{-3}	626×10^{-3}	650×10^{-3}

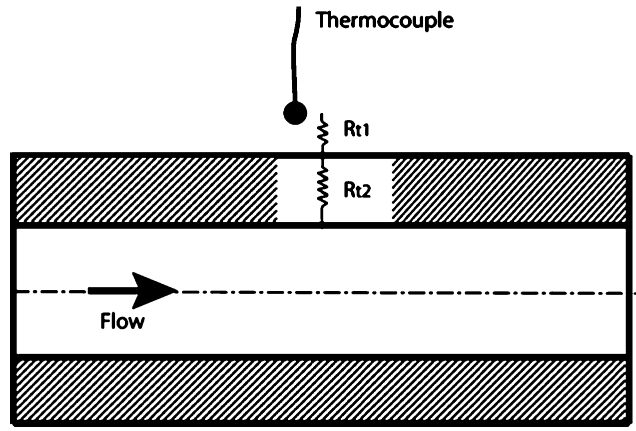

Fig. 5 Representation of the thermal resistance between the thermocouple and the flow

The conducted calibration procedure consisted in the determination of R_t for each one of the five temperature measurement stations and the obtained calibration constants will be applied to the corresponding temperature measurements made under the remaining experiments with different nanofluids. Figure 6 shows the calibrated curve for distilled water, for the Nusselt number as a function of the inverse Graetz number (Gz^{-1}), at two distinct Reynolds number.

3 Results and discussion

The experimental Nusselt number enhancement ratio, along the axial position are summarised in Figs. 7 and 8. These results suggest that the MWCNTs dispersed in DW significantly enhance the Nusselt number and therefore an enhancement to the convective heat transfer coefficient of the mixture is expected. For Re = 1,650, the results shows an average Nusselt number enhancement of 23 and 10 % respectively, at the entry and exit regions, for the 0.25 % vol MWCNT/DW nanofluid.

From Fig. 8 presents the Nusselt number enhancement variation along the test section for $Re = 2{,}060$. The obtained heat transfer enhancement for the 0.5 % vol MWCNT/DW was 47 and 44 %, for the entry and exit regions, respectively. As expected, it was verified a significantly increase in the Nusselt number with a higher flow rate.

In addition, the results suggest a higher enhancement of the Nusselt number than that observed to thermal conductivity. Such behaviour suggests that the heat transfer of nanofluids may be governed by other mechanism, different from the conductivity. Similar results were obtained by other researchers [30, 36, 52]. For instance, Xuan et al. [36] reported an 60 % enhancement of convective heat transfer coefficient and 12.5 % to the thermal conductivity, for

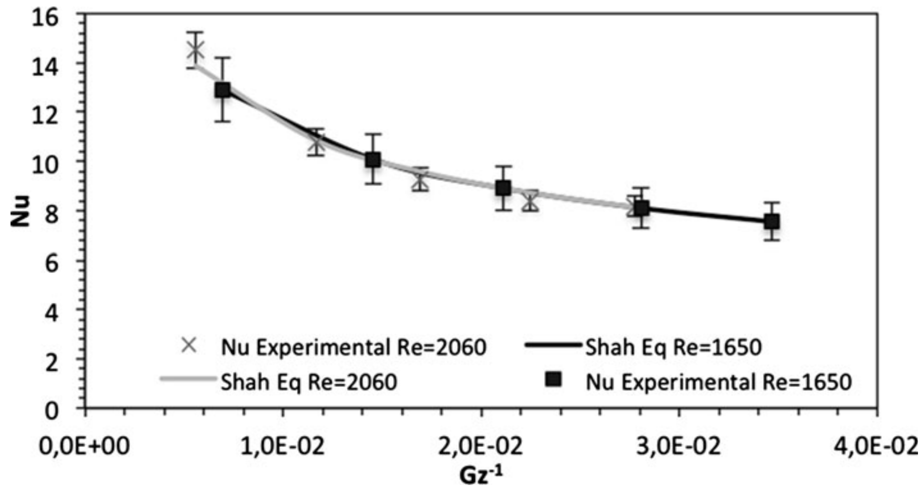

Fig. 6 Experimental calibration curve for DW

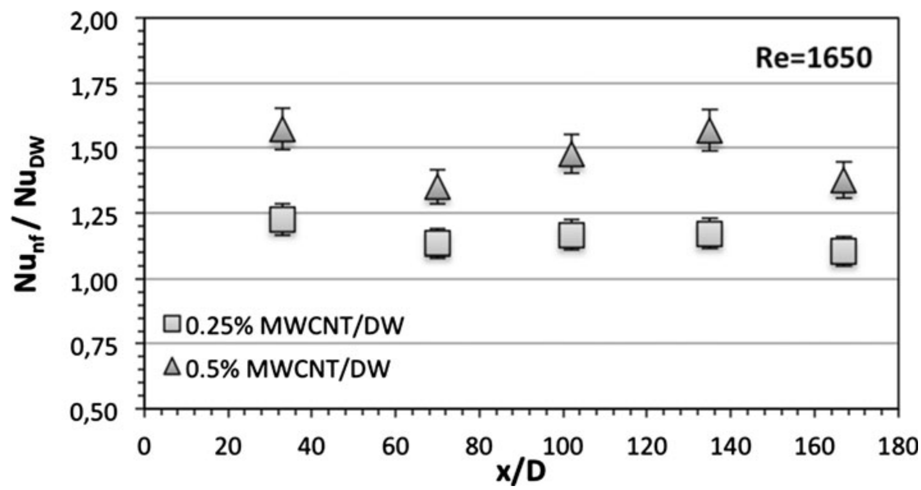

Fig. 7 Nusselt number enhancement for the studied fluids (deionised water, 0.25 % vol MWCNT/DW and 0.5 % vol MWCNT/DW), under a $Re = 1{,}650$, at different axial points

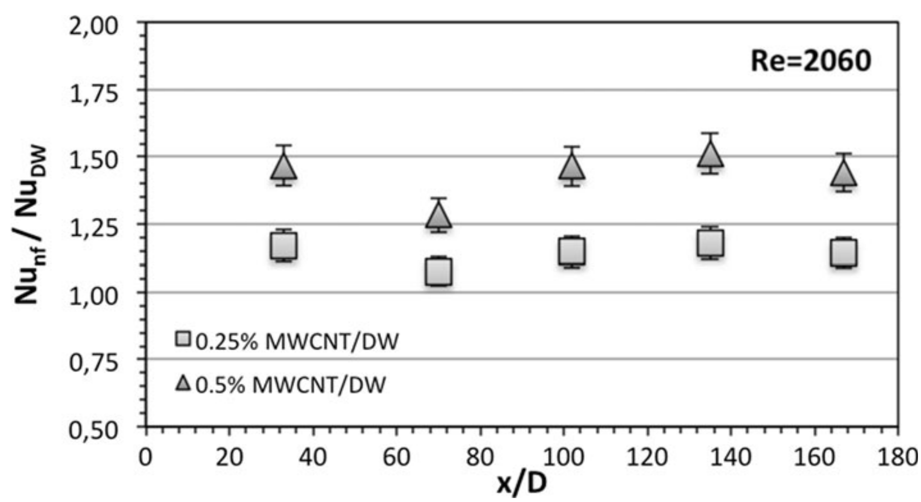

Fig. 8 Nusselt number enhancement for the studied fluids (deionised water, 0.25 % vol MWCNT/DW and 0.5 % vol MWCNT/DW), under a $Re = 2{,}060$, at different axial points

aqueous suspensions of Cu nanoparticles. Wen et al. [30], obtained an 47 % enhancement on the convective heat transfer coefficient, and 10 % enhancement on the thermal conductivity. Despite the mechanisms responsible for these heat transfer enhancements are not yet fully understood, both authors suggested that the Brownian motion of the nanoparticles might have to be considered. Nevertheless, this issue continues to be object of discussion within the

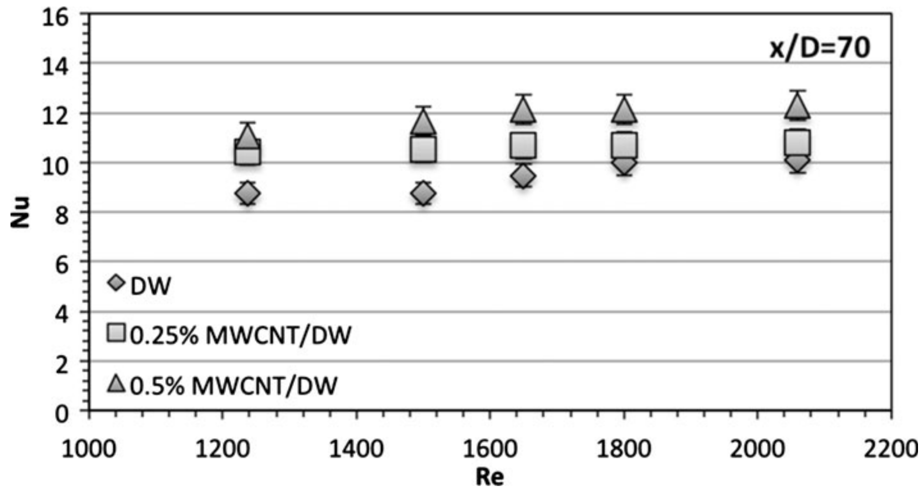

Fig. 9 Nusselt number versus Reynolds number for the DW, 0.25 % vol MWCNT/DW and 0.5 % vol MWCNT/DW nanofluids at $x/D = 70$

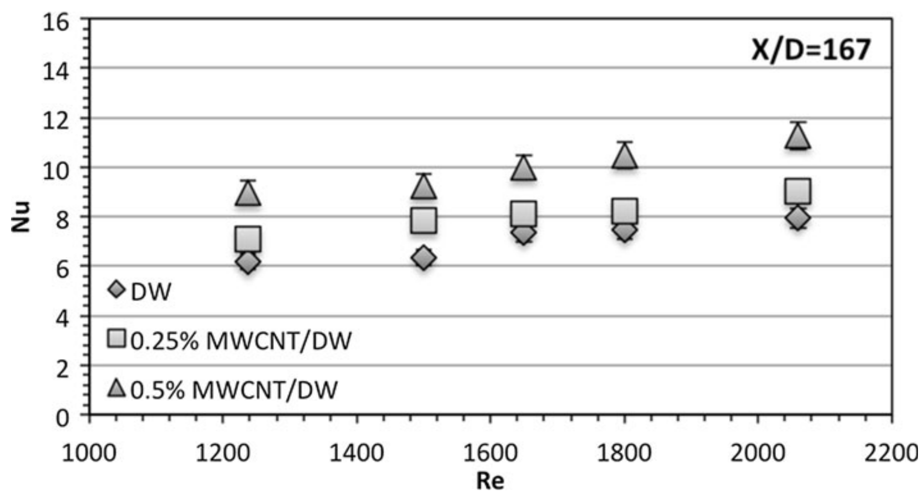

Fig. 10 Nusselt number versus Reynolds number for the DW, 0.25 % vol MWCNT/DW and 0.5 % vol MWCNT/DW nanofluids at $x/D = 167$

scientific community, and the main reasons pointed out are: nanoparticle shape, size, aspect ratio and alignment, the reduction of the boundary layer thickness, thermal conductivity enhancement and the formation of percolation structures.

In Figs. 9 and 10, a comparison for the Nusselt number as function of Reynolds number after 70 and 167 diameters from the inlet section ($x/D = 70$ and $x/D = 167$) is presented. It can be observed that as expected, the Nu rises with Re both for DW and for the analysed DW based nanofluids, however the Nu enhancement rate with the increase of Re is lower for nanofluids. This suggests that the MWCNT effect is particularly noticed at low Re. This is an intriguing behaviour that advocate for further experimental and theoretical research on the mechanisms behind the heat transfer enhancement of nanofluids.

4 Conclusions

An experimental apparatus was assembled to study the heat transfer characteristics of nanofluids. To accomplish the latter, two distinct nanofluids were developed and their thermo-physical properties measured. For a 0.5 % volume fraction of MWCNT dispersed in DW, the thermal conductivity shows an enhancement of 7.4 %. Nevertheless, the convection heat transfer experiments suggest a Nusselt number and consequently a convective heat transfer enhancement of 47 %. The experimentally obtained results suggest that nanofluids effective thermal conductivity enhancement, by itself, cannot fully explain the enhancement on the respective dynamic heat transfer capabilities. Similar results can be found in the literature for different nanofluids, and the authors suggested several mechanisms that could explain such behaviour, namely interparticle interaction and the formation of percolation networks. Therefore, this study highlights the importance of future experimental and theoretical research on MWCNT nanofluids.

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