Towards convective heat transfer enhancement: surface modification, characterization and measurement techniques

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Abstract. In this work, heat transfer surface modification and heat transfer measurement technique is developed. Heat transfer investigation was aimed to study the effect of carbon nanofibers (extremely high thermal conductive material) on the enhancement level in heat transfer. Synthesis of these carbon nanostructures is achieved using thermal catalytic chemical vapor deposition process (TCCVD) on a 50 µm pure nickel (Ni270) wire. The micro wire samples covered with CNF layers were subjected to a uniform flow from a nozzle. Heat transfer measurement was achieved by a controlled heat dissipation through the micro wire to attain a constant temperature during the flow. This measurement technique is adopted from hot wire anemometry calibration method. Synthesis of carbon nanostructures, heat transfer surface characterization and measurement technique are evaluated. Preliminary results indicate that an average enhancement in Nusselt Number of 17% is achieved.

1. Introduction

Ever since the milestone discovery of multi wall carbon nanotubes (CNTs) by Iijima [1], it was anticipated that these nanostructures would have truly remarkable mechanical and heat-transport properties, given to the strength of the carbon-carbon bond within graphene layers in graphite. Nowadays, there is growing evidence, coming from both experimental and theoretical studies that CNFs do indeed have an outstandingly high young’s modulus, high thermal stability and thermal conductivity. Due to their extremely high thermal conductivity, carbon nanotubes are considered in heat transfer researches [2-5]. The thermal conductivity at room temperature along the axis of the CNTs was found to be greater than 3000W/m.K [6] which results in a substantial improvement of the exchange of heat between the surface and the surrounding fluid flow. In contrary, the thermal conductivity at room temperature of CNTs across the axis (radial direction) is poor with a value of 1.52W/m.K [7]. Consequently, the structural arrangement graphene layers have a tremendous influence on the thermal properties. Carbon nanostructures exist in three different graphene arrangement: perfect cylindrical arrangement of graphene sheets (Tubes: CNFs), conical arrangement of the graphene sheet (fish-bone: CNFs) and flat graphene arrangement (stacked: CNFs). These different structures are believed to have different thermal conductivities.

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Figure 1. foam material used as a regenerator surface in thermo acoustic heat pumps. The metallic foam is an interconnected network cellular structure consisting of solid metals containing large volume fraction of gas filled pores. To accurately investigate the effect of CNFs on the heat transfer surfaces, geometrical simplification of the regenerator surface is made by looking into the cross-section individual strands and size. A 50 micrometer cylindrical wire surface is used for investigation.

Kordas et.al [8] have reported an efficient chip cooling (about ~1W more power was dissipated) by integrating laser patterned CNT micro fin structures on a silicon chip on 1mm² surface compared to bare chip. It was also reported that 11% of more power dissipated during the natural convection and 19% under forced convection. Zhimin Mo et.al [9] also reported an effective cooling for microelectronic applications using integrated CNT fins made by lithographic technique and TCCVD on microchannel surface. It was mentioned that the flow rates were decreased by 12% whereas the heating power input is increased by 23% keeping the transistor temperature 6°C below the reference cooler. It was also suggested that self-aligned CNTs would increase the heat transfer higher than what was achieved. However, both literatures lack appropriate method of quantifying the heat transfer phenomena.

Preliminary experimental research has shown that the presence of carbon-nano-fibers existing on a metallic foam surface can enhance heat transfer up to 40 % [10]. These fibres influence the fluid flow, and enlarge the heat exchanging surface. However, quantifying the impact of the CNF layer on the heat transfer process is challenging due to the contribution of the pressure drop on the enhancement level.

In this work, heat transfer surface modification was achieved. Carbon nano fibers were synthesized and characterized for their physical appearance (morphology) and chemical quality (crystallinity). Heat transfer measurement setup was designed to optimize and to study the impact of different morphological and topological behavior of the CNF structures used to modify the heat transfer surfaces. During the course of this paper a test measurement and analysis was made for 50µm sample Ni wire and CNFs layer deposited on similar surface and comparison was made.

2. Materials
Polycrystalline Ni wire (99.9%, Ni270), made by wire drawing mechanism, with a uniform diameter of 50µm was used in this study to represent a differential strand of common regenerator element such as metallic foam, see figure 1. The surface of the wire was modified by depositing carbon nano structural layer. High purity gases were used during the synthesis process: hydrogen (99.999%,INDUGAS), nitrogen (99.999% INDUGAS) and ethylene (99.95% PRAXAIR).
3. Experiments

3.1. CNFs layer synthesis
Prior to CNF-synthesis, the Nickel micro wire samples were pretreated under reducing atmosphere in a vertical quartz reactor which was heated from outside by an electrical furnace. The samples were heated to 600°C with a heating ramp of 6°C/min under nitrogen (inert medium) stream with total flow rate of 100 ml/min. After reaching 600°C, 30 vol.% hydrogen was introduced in the nitrogen stream to reduce the samples for 1 h while maintaining the total flow rate at 100 ml/min. This pretreatment is essential for creating grains on the nickel wire surface, which support the nucleation and hence the growth of CNFs. Followed by this reduction pretreatment, samples were flushed by with 100ml/min N2 for 30 min. The samples were further exposed to a reactive gas mixture of 20 vol.% C2H4, 5 vol.% H2 and 75 vol.% N2 at 600°C with total flow rate of 100ml/min for 1hr. Afterwards, the samples were cooled down in N2 to room temperature. Finally, samples were further exposed to a jet of air in order to remove the loosely attached carbon nano fibers.

3.2. Characterizations of CNFs layer
CNFs layers were analyzed with different characterization techniques. The morphological characteristics of the sample were studied using scanning electron microscopy (SEM; JEOL-Neoscope JCM-5000 equipped with thermally assisted field emission gun operating at 5kV and 10kV). The micro wire sample with the synthesized CNF layer was mounted on an aluminium stub by a
conductive carbon tape. Images taken were used to characterized the morphological effect of the element.

In order to quantify the surface roughness, uniformity of growth and mean diameter of the samples, the SEM image was further processed using a MatLab image processing module. The SEM image is scaled for physical dimension which was used to relate the pixels of the image to a physical dimension. Diameter distribution of the sample wire (Ni270+CNF) was analyzed and the average diameter was calculated, see figure 2. This helps to determine and decide the CNF growth time during the synthesis process.

Raman analysis was performed using Senterra Raman spectrometer (Bruker Optik GmbH) to probe the crystallinity of the CNF layer which helps to determine the alignment of the graphene planes. The graphene plane arrangement influences the conductivity of the CNFs produced by analysing the defects. The spectrometer was equipped with a CCD detector cooled at 208K (-65°C). Raman scattering was conducted in the backscattering configuration with a green laser (wavelength = 532 nm) at room temperature. The Raman spectrometer was equipped with an optical microscope, which was used to focus the incident laser and record the illuminated area. The laser beam used a microscope objective of ×50. The spectral resolution was ~9-15 cm⁻¹ and the optical power at the sample was maintained at 5 mW. Spectra were recorded for an integration time of two seconds and averaged over ten scans to improve the signal-to-noise ratio. The sample was analysed without any sample treatment or preparation.

![Raman spectrometric analysis of CNFs sample](image)

**Figure 3.** Raman spectrometric analysis of CNFs sample

3.3. Heat transfer measurement setup

The experimental apparatus is divided into two distinct parts: probe calibration and constant temperature heat transfer measurement. The schematic diagram of the heat transfer setup is shown in figure 4. Air was supplied from a 6 bar air supply and the flow was controlled with a mass flow controller. The air passes through flow straightener and reaches the nozzle. The nozzle was designed to achieve a constant velocity profile at the exit and the speed was measured using a water column. Sample wires were spot welded into a probe and placed perpendicularly facing the flow. The probe was electrically heated to a desired temperature. The temperature was controlled by a half bridge circuit. The source current was supplied and regulated by high precision DC power supply (Tektronix PWS4205). The amount of voltage drop and the resistance were measured using a NI-PCI-6280 module. The sample is made up of high purity (99.9%Ni) polycrystalline nickel micro wire (Ni270)
which is also used as a catalyst and substrate material during the synthesis of the CNF layer. This material has a high thermal coefficient of resistance which enables to detect and measure the resistance of the probe. Thermal coefficients of resistance was specified assuming a linear relationship between resistance and temperature difference. During the measurement the thermal coefficient of resistance was found to be $\sim 0.006/\degree C$. Energy-dispersive X-ray spectroscope (EDX) was used to analyse the elemental analysis of the nickel sample core after the synthesis of the CNFs layer was made. This helps to check whether carbon diffuses in to the core of the nickel wire. Results show no carbon penetration in to the nickel core. This is because of the diffusion rate of carbon in to the nickel is much smaller than the CNF growth kinetics. As a result, the thermal coefficient of resistance continues to be the same before and after synthesis.

The majority of the heat dissipated from the wire to the surrounding is mainly the result of natural and forced convection. The effect of radiation is neglected. The porosity and permeability are very important parameters to anticipate the amount of flow penetration. Due to the curvature effect of the cylinder, it is expected that both porosity and permeability of the CNFs increases along the radial direction. As a result, the characteristic diameter chosen for the non-dimensional analysis is taken to be the same as the bare wire. The Nusslet number and Reynolds number are calculated as:

$$Nu_d = \frac{l^2 R_w}{k_f \pi \nu (T_w - T_\infty)}$$  \hspace{1cm} (1)

$$Re_d = \frac{\rho_f u_{\infty} d}{\mu_f}$$  \hspace{1cm} (2)
Figure 5. Comparison of heat transfer measurement for 50µm Ni wire sample with literature values (king et al.[11] and Parnas et al.[12]).

Heat transfer measurement of the bare wire was made. Several measurements were made to evaluate the reproducibility of the heat transfer measurement and satisfactory results are obtained with an average deviation less than 2.5%. Moreover, measurement results and literature data are well-matched, see figure 5.

4. Results and discussion

SEM results show that the samples are heavily populated by CNFs. Shiny nickel particles are observed on the surface of the CNF layer. This shiny particles are nickel particles that are created during the pre-treatment process hence developed to tip type growth of CNFs [13,14] which consumes or reduce the nickel diameter hence increases the resistance of the sample during measurement. Alignment of CNFs was not achieved during the synthesis. As a result, uniform growth of CNFs layer across the length was difficult to achieve, see figure 2. High surface irregularities creates high surface roughness on the top surface of the layer, which is believed to influence the heat transfer process. As a result, accurate sample size measurement was done by an image post-processing technique using MatLab, as mentioned in section 3.2. Samples investigated after the synthesis process exhibited an average diameter of 152µm (Ni micro wire covered with CNF layer), which is equal to an average CNF layer thickness of 51µm.

The Raman spectrum of CNF layer synthesized on Ni wire is shown in figure 3. Two clear bands are visible, centered at ~1340 and ~1592 cm\(^{-1}\). It is known that the Raman spectrum of single-crystal graphite, as well as of highly oriented pyrolytic graphite (HOPG) have a single band at ~1582 cm\(^{-1}\) (G-band), which is known as graphite mode. Carbon materials with less order of crystallinity exhibit a band at ~1350 cm\(^{-1}\), which is a defect induced Raman band named as the defect mode (D-band) [15, 16]. The ratio (R) of the relative intensity of the D-band and G-band (R=I\(_D\)/I\(_G\)) can be utilized to assess the degree of graphitization and the alignment of the graphene planes. The R-value of the CNF layer synthesized on Ni wire is 1.7, which indicates the presence of interstitial defects or quasi-crystalline nature of the CNFs in agreement with literature [17]. It is known that the pyrolysis of the carbon nano-
The comparison of average Nu as a function of Re between the bare and the CNF nickel coated wire reveals that for all test cases the experimentally determined Nu number is higher for CNF coated wire compared to that of the bare wire, see figure 6(a). The average enhancement is found to be 17%. However, the exact characteristic dimension is difficult to obtain due to unknown the flow penetration area on the CNF layer. In addition, though the CNF layer has a very high porosity, the permeability is not known and it is impossible to predict the amount of flow passing through the CNF layer and the depth of the flow penetration.

Figure 6. Heat transfer measurement comparison of (a) Q/L vs. V and (b) Nu vs. Re for both bare and CNF coated wire surface. Results indicate an enhancement in both Q/L and Nu over the entire range of flow.
The amount of heat dissipated per unit length (Q/L) at different flow conditions for CNF coated wire was also compared to that of the bare wire, see figure 6(b). Result show that on average 22% of enhancement was obtained. This enhancement is possibly attributed to the enhanced area provided by the layer of CNFs and the surface roughness the CNF layer create. However, further study need to be done in determining the influences.

5. Conclusion
Synthesis of densely populated porous layer of CNF is achieved on a 50µm nickel wire using TCCVD. The produced layer is strongly attached to the nickel surface. Heat transfer measurement shows satisfactory reproducibility of the measurement results. As a result, the measurement technique is reliable for investigating heat transfer measurement on micro-scale. Preliminary heat transfer measurement results show that the modified surface has a higher heat transfer rate compared to the bare wire. However, the CNF sample quality should be improved for better crystallinity and uniformity by tailoring the experimental procedures and parameters during growth process.

6. References
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