



## Research paper

# Experimental study of the sintered multi-walled carbon nanotube/copper microstructures for boiling heat transfer



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## HIGHLIGHTS

- Influences of MWCNTs contents and coating method were studied.
- The heat transfer performance improved as the content increased up to 20 vol.%.
- A worst boiling performance was observed from the 30 vol.% MWCNT/Cu coating.
- A better enhancement was performed by double-layer coating.

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## ABSTRACT

Due to the mesh surface and intrinsic thermal conductivity of multiwalled carbon nanotubes (MWCNTs), a variety of MWCNT growth techniques are applied to boiling heat transfer enhancement. In this study Cu powders and MWCNTs were ball milled to produce composite powders. These composite powders were applied on Cu plate substrates by electrostatic spraying and were consolidated by sintering in an electric furnace. The resulting surface coatings that were fabricated from the MWCNT/Cu composite powders were considered and studied for the enhancement of the boiling heat transfer under a pool of saturated R134a. The thermophysical properties of the surface coatings were investigated. It was found that the pore density was a major factor that influenced the boiling heat transfer performance. The 20 vol.% MWCNT/Cu coating exhibited the best boiling performance. The single-layer coating was found to be less efficient in its boiling performance as compared to the double-layer coating due to the lack of the mechanical support from the Cu powder based layer.

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## 1. Introduction

Enhancement of boiling heat transfer is considered as an important way to improve the energy efficiency of refrigeration and air-conditioning systems and obtain economic benefits from these systems [1,2]. There are three material surface factors that could affect boiling heat transfer performance. These include roughness, wettability, and porosity. The presence of a porous layer on the material surface has been considered as the most important factor by far after conducting detailed investigations as reported in Refs. [3–8]. In the study reported in Ref. [9], boiling performance on five different surfaces was investigated and the results indicated that, due to the dramatically increased nucleation site density, the micro-surface produced astounding pool boiling enhancement.

Particularly, an approximate 330% reduction of initial superheat was exhibited by these surfaces. Ref. [10] reported a boiling heat transfer experiment using foam-coated tubes. A better performance was observed on the tube with higher porosity, and the enhancement was more significant on the tube with high pore density.

Recently, carbon nanotubes (CNTs) are used in numerous engineering applications due to their excellent thermal, mechanical, and electrical properties. Especially, CNTs are known to have extremely high thermal conductivity [11,12]. In the thermal management field, they have been studied and applied in surface coatings technology to improve pool boiling heat transfer coefficients. As investigated in Refs. [13–16], surfaces that were coated with multi-walled CNTs (MWCNTs) exhibited reduced incipient boiling superheat and enhanced nucleate boiling heat transfer coefficient as compared to smooth surfaces. These advantages were attributed to the mesh structure of the MWCNTs, which remarkably increased the number of nucleation sites and resulted

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in an effective initiation of the nucleation process. In addition, some studies have already indicated that the surface wettability is another important parameter influencing the boiling heat transfer performance [17–22]. Ref. [17] reported the influence of the wettability on bubble growth and departure and showed that bubbles appeared on the surface at lower heat fluxes in the case of hydrophobic surfaces. In contrast, a hydrophilic surface increases the critical heat flux (CHF) values in boiling heat transfer [23]. However, these previous studies generally used water as the working fluid. In the present study, the wettability of coatings with CNTs was investigated using R134a refrigerant.

In other previous studies [13–16], CNTs were usually coated on substrates using chemical vapor deposition (CVD) method, which made the process more complicated and hard to commercialize in the industry. In the current study, the CNTs were embedded in Cu powder particles by ball milling (BM) or mechanical alloying (MA) and then coated on Cu plate substrates by sintering. Compared with CVD method, the coating method studied in this work is easier to control and is available for mass production to reduce fabrication cost. An optimal porous coating is expected to have high pore site density and high thermal conductivity to reduce superheat temperature. The influence of coatings with MWCNT on boiling heat transfer was investigated under a pool of saturated R134a. The morphology of the MWCNT/Cu composite powders and the characteristics of the coating surfaces such as pore properties, thermal conductivity, and wettability were also investigated. The influences of the coating methods and the content of MWCNT on boiling heat transfer were studied by analyzing the effects of the surface characteristics.

## 2. Materials and methods

### 2.1. Materials

MWCNT/Cu composite powders were produced by MA using an attrition ball mill (ATM-1K, KM TECH, Korea). This process is illustrated as Step 1 in Fig. 1. The MWCNTs (diameter = 5–20 nm, length ~ 10  $\mu\text{m}$ , aspect ratio >500) were purchased from Carbon Nano-Material Technology Co., Ltd., Korea. The density and the thermal conductivity of the MWCNTs were considered to be 2.1 g/cm<sup>3</sup> and 750 W/m K, respectively. The Cu powder (ACU-325, 325 mesh, and purity  $\geq 99.5\%$ ) was purchased from Chang Sung Corp., Korea. The MWCNT and Cu powder, together with zirconia milling balls (5 mm), were placed in a zirconia milling jar. The ball-to-powder weight ratio (BPR) was 20:1. The jar was then filled with argon and sealed to keep an inert environment during the milling process. To prevent the powders from heating too much during milling, cooling water was supplied into the milling jar of the attrition ball mill. The milling speed was 900 rpm and the milling time was 4 h.

### 2.2. Fabrication of the surface coatings

The fabrication of the surface coatings is illustrated as Step 2 in Fig. 1. In this step, a prepared 2 wt.% polyvinyl alcohol (PVA) solution was sprayed on the cleaned surface of a Cu plate substrate. This PVA solution served as adhesive that facilitated the adhesion of the powder particles on the substrate during electrostatic spray coating (Step 3). As shown in Fig. 1, Step 3 has two methods indicated as Steps 3–1 and 3–2. For Step 3–1, the coating was only single-layer and was made from the MWCNT/Cu composite powder. This is referred as the single-layer coating method. On the other hand for Step 3–2, referred as the double-layer coating method, the coating was fabricated with two layers. The first layer (Step 3–2a) was made from pure Cu and the second layer (Step 3–2b) was made

from the MWCNT/Cu composite powder. The coating of pure Cu powder was done in order to increase the porosity of the coatings and aid the adhesion of MWCNT/Cu composite powder on the surface. Step 2 was repeated before Step 3–2b. The coating thickness was controlled by directing the flow of powder using an electrostatic spray gun and controlling the deposition rate. The thickness of all the coatings ranged from 90 to 100  $\mu\text{m}$ .

Sintering was performed as Step 4 in Fig. 1. In this step, the coated Cu plates were placed inside an electric furnace. The temperature was raised from room temperature to 350  $^{\circ}\text{C}$  at a rate of 20  $^{\circ}\text{C}/\text{min}$  and held for 10 min in an air atmosphere to remove the PVA. The temperature of 350  $^{\circ}\text{C}$  was considerably lower than the MWCNT oxidation temperature, which generally would begin at 400–430  $^{\circ}\text{C}$  [24,25]. The temperature was further raised up to 600  $^{\circ}\text{C}$  and held for one hour in a mixed gas (hydrogen 10%, nitrogen 90%) environment for sintering and reduction. Then the furnace was cooled down to room temperature.

Several sintered and coated Cu plate specimens were prepared with different parameters. These specimens are summarized in Table 1.

### 2.3. Characterization

The particle diameters of the composite powders ( $d$ ) were measured using an optical microscope and image processing software (IMT I-Solution, IMT i-Solution Inc., Canada). The morphologies of the powders and coatings were examined by field emission scanning electron microscopy (FESEM, S-4800, Hitachi, Japan).

For the coatings, quantitative examination was performed by mercury porosimetry (Auto Pore IV 9500, Micromeritics Instrument Corp., USA) to determine the porosities ( $\varepsilon$ ) and the average pore diameter ( $D$ ) of the coatings. The pore size was determined based on the external pressure needed to force mercury into a pore and on the liquid surface tension. The pressure range was 0–413.7 MPa and the pore size range was 0.003–360  $\mu\text{m}$  in diameter. The intrusion resolution was 0.1 L/g and the intrusion accuracy was 0.1% of the intrusion volume. The pore density is defined as the quantity of pore in a 1 cm<sup>3</sup> volume of coating. Pore density of each of the enhanced surfaces was calculated using Eq. (1):

$$\rho = \frac{\varepsilon}{V} \quad (1)$$

where  $\rho$ ,  $\varepsilon$ , and  $V$  were the pore density, porosity, and individual pore volume, respectively. For simplicity, the pore shape was assumed to be spherical. Thus, the volume of individual pore was simply calculated as

$$V = \frac{\pi D^3}{6} \quad (2)$$

To assess the wettability of the surfaces due to the thermodynamic properties of R134a, the contact angle (CA) values between the surfaces and liquid refrigerant were determined by measuring the height of capillary rise on vertical flat plates. The CAs were calculated using Eq. (3) [26]:

$$\sin \theta = 1 - \frac{\Delta \rho g h^2}{2\gamma_{lv}} \quad (3)$$

where  $\theta$  was the contact angle of the capillary rise profile on the vertical plate;  $\Delta \rho$  was the density difference between the liquid and the vapor phases of the refrigerant;  $g$  was the acceleration due to gravity;  $h$  was the height of capillary rise; and  $\gamma_{lv}$  was the surface

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