Condensation heat transfer enhancement by surface modification on a monolithic copper heat sink

Ding-Jun Huang a,c, *, Tzong-Shyng Leu b,c

a Department of Materials Science & Engineering, National Cheng Kung University, Tainan 70101, Taiwan, ROC
b Department of Aeronautics & Astronautics, National Cheng Kung University, Tainan 70101, Taiwan, ROC
c Institute of Nanotechnology & Microsystems Engineering, National Cheng Kung University, Tainan 70101, Taiwan, ROC

HIGHLIGHTS

- Superhydrophobic surface modification is applied to a monolithic copper heat sink.
- A monolithic copper heat sink is used to prevent contact thermal resistance.
- EGC-1720 fluorosilane polymer is employed as the waterproof agent.
- Durability of the EGC-1720 coated surface is investigated.
- The relative heat transfer enhancement of the heat sink is compared.

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ABSTRACT

In this study, the condensation heat transfer performance on a pure copper surface, as well as a superhydrophobic-modified copper surface were compared. Differing from other condensation heat transfer experimental designs, a monolithic copper heat sink was utilized in this study to prevent contact thermal resistance and/or thermal conduction limitation of the thermal paste applied between the modified condensation surface and heat sink plate. This approach has not yet been documented in the literature. The superhydrophobic copper heat sink surface was prepared using a hydrogen peroxide immersion and fluorosilane polymer (EGC-1720) spin-coating. Experimental results show that the condensation heat transfer performance on the superhydrophobic copper surface is superior to that of a pure copper surface. Additionally, durability tests of the pure and superhydrophobic coating copper surfaces in a harsh vapor environment are discussed in this study.

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1. Introduction

The wettability of solid surfaces is a very important aspect of materials science and surface chemistry. In recent years, surface modification to change the wettability of a material’s surface has become a promising technique and attracted much attention. Generally, surface modification techniques play an important role in many practical and industrial applications, such as self-cleaning [1,2], anti-icing and -frosting [3–5], oil-water separation [6–8], drag-reduction [9], corrosion resistance [10], and condensation heat transfer (CHT), among others.

CHT refers to the heat transfer process when a phase change occurs on a subcooled surface under a vapor environment. Vapor condensates on the subcooled surface and transfers a large amount of latent heat energy onto the surface of the material. Depending on the surface wettability of the material, condensation can be divided into two modes: “dropwise condensation” (DWC) and “filmwise condensation” (FWC). DWC forms sphere-like droplets on hydrophobic surfaces, while FWC appears as a film-like liquid layer on hydrophilic surfaces. DWC and FWC surface nucleation modes have considerable influence on CHT performance. Typically, the CHT performance of DWC outperforms that of FWC because DWC accelerates the processes of condensation, nucleation, droplet-growth, coalescence and droplet-falling per unit time.

To ensure optimal performance of CHT, material selection is important. In this regard, copper [11–13], with its excellent thermal conductivity [14,15], has become one of the most important materials in CHT devices. Many copper surface modification...
techniques have been reported and applied in CHT experiments [15–25]. Edwards & Doolittle [16] developed a copper tube coated with Tetrafluoroethylene (Teflon) and found that it enabled better CHT efficiency than without the coating. Also, comparisons among different coating films were made, which demonstrated that differences between heat transfer coefficients and coating film lifetime among certain surface coatings influenced CHT performance. A similar issue was investigated by Marto et al. [26], who adapted various surface coating layers to coat different metal tubes and studied the heat transfer performance and endurance at low pressure. Their results indicated that copper and copper–nickel alloys achieved higher heat transfer coefficients when coated with silver; however, durability of the silver coating under a vapor environment was limited compared to other organic coatings like Parylene-D, No-stick or fluoroacrylic. They found that although the copper coated by No-Stick was more durable, its heat transfer performance was inferior to that of the silver coated copper surface. Clearly, heat transfer performance and coating longevity must be studied and tradeoffs optimized. Holden et al. [27] extended the above results by visually verifying the condensate droplets for surface coating endurance and longevity, the results of which can be used as a basic reference for optimization.

As a result, a research on superhydrophobic coating agents mentioned above, some research results also have used organic self-assembled monolayers (SAMs) as a coating agent for surface modification. Das et al. [22] employed hexadecylthiol to coat the surfaces of copper and other alloys, and found that a SAM on copper or copper alloys was able to achieve superior heat transfer performance to that on gold-coated aluminum or oxidized aluminum. They also investigated these materials by controlling the chamber pressures, and documented high CHT performance of SAMs-coated copper and copper alloys. It still remained true that DWC enhanced heat transfer more at various levels than FWC. In a related study, Vemuri et al. [23] investigated not only SAM coatings, but also bonding strengths with respect to CHT. They examined DWC by using an n-octadecyl mercaptan SAM-coated copper tube and a stearic acid SAM-coated one. Because the n-octadecyl mercaptan SAM layer reduced the cupric oxide layer on the copper surface to form a uniform organic film of closely packed chains of thiols, durable covalent bonding was achieved. According to their findings, the durability of the covalent bonding was superior to the hydrogen bonding formed by the stearic acid SAM layer. The CHT enhancement of the n-octadecyl mercaptan-coated SAM tube was twice as large as that of the heat transfer coefficient of their hydrophilic copper tube, even when the experimental time exceeded 2000 h. Subsequently, Chen et al. [28] used a similar modification method as Vemuri et al. [23,24] for investigating the DWC performance of a copper block condenser. The heat transfer performance was also good due to the bonding strength between the SAM layer and copper surface, thereby extending the lifetime. The average convection heat transfer coefficients of the DWC vertical surfaces were 1.7–2.1 times greater than those of FWC.

Surface roughness is also a factor that influences DWC performance because it is required in fabricating a hydrophobic surface. For example, Izumi et al. [25] investigated the heat transfer characteristics of DWC on a surface with round shaped grooves coated with hydrophobic oleic-acid. The heat transfer performance was clearly enhanced by optimizing groove widths, which indicates that performance improvements may be achieved by tuning surface roughness. It has been suggested that further enhancements could be expected if two-tiered nanostructures [29,30] were fabricated on the grooves to form a superhydrophobic surface. In the aforementioned study [25], a heat sink-like plate was utilized as the experimental sample, and its feature seemed like a monolithic heat sink bulk. It is well-known that a monolithic substrate is able to prevent thermal resistance between the condensation side and the cooling side; as such, it is preferred in experiments or practical applications, and is commonly adopted in CHT studies. For example, Ma et al. [21] investigated CHT enhancement in the presence of non-condensable gas (NCG), and found that the lower the amount of NCG, the higher the heat transfer coefficient. In addition, their condensing block was similar to a monolithic copper heat sink, so it was a better reference for the choice of experimental substrate. Lan et al. [19] also utilized a similar monolithic condensing block to investigate the CHT performance of a surface with two fabrication processes, but the both featuring the same SAM coating. Through the fabrication processes, they obtained a superhydrophobic surface and a hydrophobic one. Surprisingly, the CHT performance of the superhydrophobic surface was lower than that of the hydrophobic surface. They stated this was because of the adhesion effect of the hierarchical micro- and nano-structures, which induced an adhesion effect that increased the droplet growth cycle time. Their article suggested that the coating and surface processing should be optimized if more heat transfer is desired. Further, Leu et al. [31] also discussed the importance of the plate material and processing features. In their study, a silicon surface was modified as a composite wettability gradient surface and then attached onto a copper heat sink. Compared with pure silicon, the heat flux of the modified silicon was increased by 10%; however, no details regarding the bonding between their modified silicon and copper heat sink were given. One could expect that a thermal contact resistance surely existed if there was no suitable connection media between the both materials. Moreover, it is possible that the heat transfer performance was limited. In addition, not only did the connection affect the heat transfer enhancement, but also the material of the silicon chip. Silicon has a lower thermal conductivity than copper, which is a shortcoming in thermal properties.

Therefore, from the aforementioned studies, CHT performance appears to be determined by correlations among many factors, such as surface structures, coating agents, gas concentration, etc. Comparatively, combining the use of rough surface structures and chemical coating layers is the general approach for manipulation, and constitutes the simplest way to investigate heat transfer enhancement by controlling the least amount of parameters. In the above, superhydrophobic surfaces were shown to achieve DWC and enhance CHT performance. However, enhancements must be analyzed to determine whether they are truly superior because different material processing conditions and experimental system setups can introduce variability.

In this study, a surface modification technique was developed and applied to the surface of a monolithic copper heat sink for application in CHT, the details of which are addressed for practical utilization. An EGC-1720 coating was utilized to modify the surface, and the heat transfer performance of the modified surface was investigated. For this application, the coating formed by the copper surface modification technique must be able to withstand high temperature vapor environments. Therefore, one of the research goals was to investigate whether the modified copper surface can be sustained under such a harsh environment.

2. Experimental process

2.1. Types of copper heat sinks

A commercial heat sink of approximately 90 × 60 × 27 (mm³) (Item No. TPCU008, Tai-Pao Int. Corp.) was utilized in our experiment, as shown in Fig. 1. The fabrication of the heat sink mainly included relief cut, planning with shaping, and formation of a monolithic part. Therefore, the contact thermal resistance between
the fins and their bottom surface could be eliminated. A monolithic copper heat sink is classified into two types in this study, as shown in Fig. 2, namely, a pure heat sink and a superhydrophobic one. The pure copper heat sink was used as received in the CHT experiment (Fig. 2(a)), while the superhydrophobic one was chemically processed and subsequently waterproof agent-coated (Fig. 2(b)). Fabrication of superhydrophobic copper heat sink follows the reported technique in Ref. [12], where modification of the copper surface was accomplished using hydrogen peroxide oxidation and Teflon coating. The process is shown in Fig. 3 and elaborated in the next section.

### 2.2. Fabrication of superhydrophobic copper heat sink

#### 2.2.1. Copper heat sink cleaning

Initially, the pure copper heat sink was immersed in a 15 wt% nitric acid (HNO₃) solution with stirring for 15 min, as shown in Fig. 3(a). This process was used to remove native oxide and impurities on the copper substrate, and is called “chemical polishing” since copper oxide and other impurities can be removed efficiently using HNO₃ solution, allowing the cleaned surface to become chemically active [11,12]. After cleaning, the copper heat sinks were washed with water, blown dry with compressed air, and heated at 60 °C on a hot plate for 10 min. During heating process, the cleaned surface was faced up to minimize any physical damage on the cleaned surface.

#### 2.2.2. Copper surface roughening

After chemical cleaning, the cleaned copper heat sink was oxidized by immersing in a 30 wt% hydrogen peroxide (H₂O₂, J. T. Baker, U. S. A.) aqueous solution with stirring, as shown in Fig. 3(b). To avoid non-uniformity in the chemical reaction, the heat sink was raised by glass supporters at both edges of the heat sink, as can be seen in the figure. A magnetic stirrer was used to accelerate the chemical reaction. The reaction area of the copper surface was larger than the effective area used in the CHT experiments. After 1 h of immersion, the reacted surface of the heat sink was then composed of CuO nanowire structures. The analytical details of the CuO nanowire structure can be found in our previous study [12]. These nanowire structures comprised the roughened layer shown in Fig. 2(b). After the H₂O₂-immersion, the copper heat sink was rinsed with water, blown dry with compressed air, and heated on a hot plate at 60 °C for 3 min. The produced nanowire structures roughened the surface to form sufficient air gaps for achieving superhydrophobicity later.

#### 2.2.3. Spin-coating

The spin-coating procedure is shown in Fig. 3(c). At the start of the procedure, an electronic liquid called EGC-1720 (Novel™ EGC-1720 Electronic Coating, 3M, U. S. A.) was prepared. This solution is composed of fluorosilane polymer and hydrofluoroether solvents, and was utilized as received without any dilution. Next, the aforementioned H₂O₂-immersed copper heat sink was placed upside-down and fixed on the acrylic cylinder. The spin-coating speed was 3000 rpm and the spin time was 20 s after pre-spinning. Finally, without any water-rinsing and air-blowing, the EGC-1720 coated heat sink was directly heated on a hot plate at 90 °C for 60 min; subsequently, the fabrication of a superhydrophobic copper heat sink was complete, as was illustrated in Fig. 2(b).

### 2.3. Condensation heat transfer (CHT) experiment

#### 2.3.1. Chamber design and part assembly

The experimental chamber is composed of one cooling chamber and one condensation chamber. In this study, the cooling chamber was designed according to the optimally designed chamber of Delsman et al. [33], the purpose of which was to enhance the water flow uniformity among the fins. The condensation chamber was used to condense vapor on the surface of the copper heat sink. For thermal insulation, the chamber material was acrylic, and chloroform was utilized to bond all acrylic parts. A 1 mL glass syringe

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**Fig. 1.** Photo and engineering draft of the monolithic copper heat sink, measurement unit in millimeters (mm).

**Fig. 2.** Types of copper heat sink: (a) pure copper heat sink, and (b) superhydrophobic copper heat sink.
(Hamilton Co. Ltd., U. S. A.) was employed to drip chloroform onto the acrylic parts, allowing the parts to be tightly assembled. After drying and evaporating any remaining chloroform, the assembled acrylic chamber was complete. Fig. 4(a) shows the cooling chamber and the installed pure copper heat sink, as well as the cooling water inlet and outlet. In Fig. 4(b), the condensation chamber is shown, which has an effective condensation surface area of approximately $34 \times 69$ (mm$^2$); the vapor inlet and outlet can be seen as well.

2.3.2. Experimental process

After the copper heat sink was installed in the chamber, all required tools and devices were connected, as shown in Fig. 5. The cooling chamber enabled flowing water to be transported by a submersible pump, with valves 1 and 2 separately responsible for controlling the cooling water supply switch. A rotameter was used as a flow meter, and was also employed to visually check the amount of cooling water and finely control the volume. The measured range was from 20 cc/min to 2000 cc/min, and three flow meters were prepared and utilized in turn. The flow meter was replaced by another one depending on the mass flow rate of the cooling water. The ranges of the three flow meters were from 20 cc/min to 200 cc/min, 100 cc/min to 1000 cc/min, and from 500 cc/min to 5000 cc/min, respectively. Deviations of the three flow meters were ±5 cc/min, ±20 cc/min and ±100 cc/min, respectively. $T_1$ and $T_2$ are J-type thermocouples and measured the temperature of the cooling water inlet and outlet, respectively. The thermocouple was connected with a thermocouple measurement device (NI USB-TC01, National Instruments, U. S. A.), the latter of which was also connected to a personal computer (PC) and had a measurement accuracy of 0.1 °C/s. The cooling water flowed through the fins and outlet, and then to the water drain tank.

The condensation chamber enabled vapor to flow freely so that heat was transferred from vapor to the condensation surface. The condensate water droplets appeared and grew on the condensation surface as the water vapor was condensing. Vapor was produced by the electric vapor generator (MBA3, Sussman-Automatic Corp., U. S. A.) and the required water for vapor-producing was supplied from the water tank by a pump. Valve 3 was used to control the vapor switch supplied to the chamber. $T_3$ is a K-type thermocouple, meaning that the vapor temperature at the vapor inlet of the chamber could be measured. $T_4$ is also a K-type thermocouple and was embedded within a drilled hole, which was 3 mm below the condensation surface, as in Ref. [34]. The reading value of $T_4$ was taken into account to calculate the condensation surface temperature of the heat sink, the calculation procedure of which is introduced in next section. $T_3$ and $T_4$ used the same measurement devices as $T_1$ and $T_2$, and all temperature measurements started when the read temperatures stabilized in their temperature plots. After surface condensation, the remaining vapor and condensate water flowed through the vapor outlet and into the drainage collector. A CCD camera was utilized to capture real time videos and photos of the surface condensation features; thus, the removable cover plate of the chamber was detached when the vapor condensation process was recording. Fig. 6 shows the
detailed conditions of the experimental chamber and connected thermocouples.

2.4. CHT theory and analysis

In this experiment, the temperature values were also denoted from the thermocouples respectively $T_1, T_2, T_3,$ and $T_4,$ as shown in Fig. 5. The volumetric flow rate was transformed as a mass flow rate assuming the cooling water density was constant. Heat transfer performance was then investigated and analyzed. The calculation and analysis of heat transfer performance were based on the results in Leu et al. [31]. Therefore, the heat flux $q$ was calculated as:

$$q = \frac{\dot{m}C_p(T_2 - T_1)}{A} \text{ (W/m}^2\text{)}$$  \hspace{1cm} (1)

where, $q$ represents the CHT heat flux on the copper heat sink surface; $A$ is the effective condensation area, which was $34 \times 69$ (mm$^2$) for each heat sink; $T_3$ and $T_4$ are the outlet and inlet temperatures of the cooling water, respectively; and, $\dot{m}$ and $C_p = 4.18$ (J/kg·C) are the mass flow rate and the constant-pressure specific heat of cooling water, respectively.

The CHT coefficient $h_c$ of the experiment was based on the results of Pang et al. [17] and Vemuri et al. [23,24], and calculated as below:

$$h_c = \frac{q}{\Delta T} = \frac{q}{T_3 - T_{surf}} \text{ (W/m}^2\text{·K)}$$  \hspace{1cm} (2)

where, $\Delta T$ is the temperature difference between the vapor temperature $T_3$ and the temperature of the condensation surface $T_{surf}.$ Here, $T_{surf}$ was found by:

$$q = -k_c \frac{dT}{dx} = -k_c \frac{T_4 - T_{surf}}{\Delta x} \quad \& \quad T_4 + \frac{q\Delta x}{k_c} = T_{surf} \text{ (°C)}$$  \hspace{1cm} (3)

Eq. (3) represents Fourier’s heat conduction law for which the 1-D thermal conduction mode is assumed; where, $T_4$ is the temperature inside the copper heat sink; $k_c$ is the thermal conductivity of copper, 401 W/m·K; and, $\Delta x = 3$ mm is the distance between Thermocouple 4 (labeled as $T_4$) and the condensation surface.

2.5. Surface characterization

The contact angles were measured using a contact angle instrument (FTÅ125, First Ten Ångstroms, U. S. A.), and the test liquid was a 4.0 mL water droplet under the ambient temperature and atmosphere. Surface morphology was assessed via scanning electron microscope (SEM, JSM6700-F, JEOL, Japan).

3. Results and discussion

3.1. Features of copper heat sink

The pure copper heat sink shown in Fig. 7 was directly utilized as received without any processing. The water contact angle (WCA) on the surface was $106.17° \pm 1.58°.$ Generally, a copper surface is slightly hydrophobic, with its wettability differing slightly depending on surface processing. To obtain a superhydrophobic
copper heat sink, a pure copper heat sink was taken into the modification process described in Sec. 2.2. After an initial cleaning by HNO₃, the cleaned surface was immersed in H₂O₂ solution. An oxidation reaction on the copper surface occurred when the cleaned copper was immersed in H₂O₂. Thus, the copper surface was transformed into a surface spread by CuO nanowire structures, whose properties were discussed in our previous study [12]. Afterwards, it was taken to be spin-coated by EGC-1720 fluorosilane polymer, as shown in Fig. 8. In Fig. 8(a), the water droplets on the surface appear spherical, and the surface was considered superhydrophobic since the WCA was 153.43° ± 5.13°. Fig. 8(b) shows the SEM photo of the CuO nanowire-structures roughened layer on the surface for achieving superhydrophobicity after spin-coating EGC-1720 on the roughened layer. These photos verified that the superhydrophobic modification of EGC-1720 spin-coating was as effective as that spin-coated by the diluted Teflon solution in our previous work [12]. Therefore, compared with the Teflon spin coating, the EGC-1720 spin coating was more convenient due to it being dilution-free, which makes it preferable for related surface coating processes.

3.2. Durability test and analysis

The durability of surface wettability always influences CHT performance. Thus, before the CHT experiments, the wettabilities of both condensation surfaces under a vapor environment according to operation time were considered, the results of which are shown in Fig. 9. In this figure, the wettabilities of both condensation surfaces were quite distinct. The WCA on the pure copper surface immediately changed from 110° to 90°, and was maintained at 90° within the operation time; however, the superhydrophobicity of the copper surface obviously degraded within a short time. The average WCAs decreased within the 6 hr operation time, but approached a constant value afterward. The average stabilized WCA was approximately 120°. According to the product information [32], the EGC-1720 coating can covalently bond to metal oxide layers, with a film coating of about 5–10 nm in thickness; however, the test results showed that the durability was inferior to that in Vemuri et al. [23]. In their study, a sample with covalent bonding between the surface and SAM coating was used, which they claimed could endure vapor condensation for at least 100 hr. In its product information [32], it is not indicated that EGC-1720 can withstand a harsh vapor environment; so possibly, the failure of the EGC-1720 coating was caused due to the continuous flushing and heating by condensate droplets and water vapor. Thus, it is speculated that the degradation of the EGC-1720 coating can be attributed to the weakened bonding between the EGC-1720 coating layer and CuO nanowire structure surface under vapor condensation conditions. Be that as it may, to the authors’ knowledge, there is no relevant experimental information or data about using EGC-1720 coating in CHT. Thus, the durability of EGC-1720 coating under a water vapor environment is just one of the
practical references for conducting the following CHT experiment in this study.

3.3. Surface condensation analysis

In addition to the durability test, surface condensation analysis was also conducted and is one of the practical references for conducting the CHT experiment. Features of the condensate droplets on the condensation surface were recorded, as shown in Figs. 10 and 11, respectively. The time labeled under each image is the corresponding operation time at the point when each image was captured. The condensation of the pure copper heat sink (as shown in Figs. 2(a) and 7) is described first, and shown in Fig. 10. From Fig. 10(a), at the starting point \( t = 0 \) s, the DWC mode appeared on the condensation surface, and many droplets of various sizes spread around the whole area. At \( t = 2 \) s, the falling droplets resulted in coalescence among other nucleating droplets along its falling path. At \( t = 4 \) s, new small droplets were nucleating again in the region cleared by the falling droplet. Afterwards, the images show that larger condensate droplets actually swept and cleared the smaller ones. At \( t = 16 \) min 0 s, as shown in Fig. 10(b), a film-like droplet appeared on the right side of image, and at \( t = 16 \) min 2 s, the width of the film-like droplet enlarged. From \( t = 16 \) min 5 s to 16 min \( t = 10 \) s, in addition to the previously mentioned film-like droplet, a variety of film-like droplets of different sizes were growing nearby. It became apparent that the hydrophobicity was obviously influenced due to the continuous flushing and heating by condensate droplets and water vapor. At 39 min operation time, as shown in Fig. 10(c), film-like droplets spread and nearly covered the entire area at all video capturing times. The profiles of the condensate droplets were not spherical, as what had appeared at the initial operation time \( t = 0 \), but rather film-like. Thus, it can also be found that FWC appeared at the condensation surface, and adhesion between the condensate droplets and condensation surface appeared as well. This adhesion resulted in the hysteresis of droplets on the condensation surface.

The condensation of the superhydrophobic copper heat sink (as shown in Figs. 2(b) and 8) is described below, and displayed in Fig. 11. From Fig. 11(a), at the initial operation time \( t = 0 \) s, DWC effects were visible on the surface and many spherical condensate droplets of different sizes appeared. At \( t = 4 \) s, coalescence occurred among the condensate droplets, and at \( t = 11 \) s, the droplets began to fall. At \( t = 17 \) s, the droplets at the bottom-middle had already fallen but some droplets still remained on the condensation surface. For a while, at \( t = 27 \) s, some new small condensate droplets nucleated and grew at the region that was cleared by the falling droplets. Nevertheless, slight adhesion between the droplets and the surface occurred, after which the droplets were not as smooth and round as those appearing on the condensation surface of the pure copper heat sink shown in Fig. 10(a). This adhesion feature is able to be explained by Narhe et al. [35], who documented that any type of condensate-droplet coalescence always results in the most stably adhered droplet (Wenzel wetting mode). Since the energy gained by the droplets during coalescence was sufficient for inducing the transition from a spherical or spherical-adhered droplet to a stably adhered droplet, the pinning effect on the droplets was increased. Thus, many non-circular droplets were observed on most of the surface area due to droplet coalescence and pinning. Another reason is that the influence was caused by the surface morphology and the EGC-1720 coating layer. Related studies [3, 4, 19, 36, 37] have indicated that hydrophobicity does not equal lower droplet contact angle hysteresis (CAH), and vice versa. However, the condensation mode was still DWC. Although adhesion between the surface and droplets was evident, the droplets on the condensation surface were still acceptably smooth and round. At the operation time of 3 h, as shown in Fig. 11(b), the profile of the condensate droplets were less round and the adhesion more obvious than that at the starting point, as shown in images \( t = 3 \) h 0 s, \( t = 3 \) h 3 s and \( t = 3 \) h 7 s, respectively. It is possible that the more irregular droplets and adhesion were influenced by the slight deterioration of the EGC-1720 coating layer. At \( t = 3 \) h 9 s and \( t = 3 \) h 13 s, the images respectively show the fall of condensate droplets and nucleation. Nevertheless, acceptable DWC mode still existed on the condensation surface at that time period.

According to Neumann et al. [38], strong adhesion of condensate droplets results in a decrease in the heat transfer performance of a superhydrophobic heat sink. Thus, the expected operation time of our superhydrophobic copper surface was controlled to within

![Fig. 10. Condensation and droplet nucleation process on the condensation surface of the pure copper heat sink. The labeled time below each image represents the operation time. (a) Starting point, (b) after 16 min, and (c) after 39 min. The yellow bar is 1.6 mm in scale. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)](image-url)
3 h for the CHT experiment. This decision was based on the results shown in Fig. 11(b) and the durability test (as shown in Fig. 9 of the previous section). Within 3 h operation time, the WCA of the superhydrophobic copper surface was approximately 140°, which still constitutes a superhydrophobic surface. In contrast, the pure copper surface underwent 2 h operation time under the vapor environment before the CHT experiment. This decision was also based on the results shown in Fig. 10 and the durability test (as shown in Fig. 9 of the previous section). This pretreatment was used to stabilize the surface wettability of the pure copper surface, so that the measurement error due to the change of wettability was minimized.

3.4. Heat transfer analysis

3.4.1. Heat transfer flux vs. temperature difference

Fig. 12(a) shows the relationship between the heat flux q and the temperature difference $\Delta T$ for both copper heat sinks. The behaviors of the heat flux q were also distinct between the FWC and DWC modes within the measured $\Delta T$ range. The heat flux of the pure copper heat sink is denoted as $q_P$. Clearly, $q_P$ was stably increasing with increasing $\Delta T$. In the CHT experiment, the pure copper condensation surface was previously operated for 2 h for steadiness (based on the result shown in Fig. 9). Afterward, the stable surface wettability resulted in stable condensation and nucleation, the trend of which was ideally increasing. Compared with the results of Ma et al. [39], the range of $q_P$ in our experiment was similar. This offers direct evidence that the results of the monolithic copper heat sink in this experiment agree with those of their customized plate, and were able to be achieved with a reasonably simple setup.

Each superhydrophobic condensation surface was operated for 3 h as its maximum operation time, after which point the used superhydrophobic heat sink was replaced with a new one. This manipulation was conducted for minimizing the deviation resulting from the degradation of superhydrophobicity (based on the result shown in Fig. 9) in each CHT experiment. The heat flux of the superhydrophobic copper heat sink was higher than that of the pure copper heat sink at the same $\Delta T$; thus, the superhydrophobic condensation surface actually enhanced the CHT performance, despite the substrates both being copper. Here, we defined the heat flux $q$ of the superhydrophobic surface as $q_{SH}$ for convenience. $q_{SH}$ increased with increasing $\Delta T$, the trend of which was like a parabola represented as a quadratic function. When $\Delta T$ was at 20 K, $q_{SH}$ was not enhanced by increasing $\Delta T$, but approached a limit instead. Thus, some explanations are given as follows: one possible reason is the limited heat transfer of the cooling water, which means that the cooling water convection could not transfer enough heat energy from the copper plate; a second possibility is the slight degradation of the EGC-1720 coating layer, even when the operation time was less than 3 h, since any coating layer degradation heavily influenced the heat transfer performance, especially when $\Delta T$ increased; and, a third reason could be according to Rausch et al.
who noted that a thermal resistance was caused by the increase in condensate mass present on the condensation surface with increasing $\Delta T$, so that the heat transfer was unable to be further enhanced.

Compared with other studies, $q_{SH}$ in this study was greater than that of Lan et al. [19], and within 20 K in $\Delta T$ of Ma et al. [39], as shown in Fig. 12(a). But when $\Delta T$ was more than 20 K, $q_{SH}$ was the least among all compared studies. Nevertheless, the superhydrophobic copper surface in this experiment behaved similarly at most $\Delta T$ ranges.

The fitting curves of $q_P$ and $q_{SH}$ in Fig. 12(a) were performed to compare the relative heat transfer enhancements of the superhydrophobic heat sink and the pure copper heat sink, for which an enhancement gain plot $\Delta q = q_{SH} - q_P$ is shown in Fig. 12(b). Increased $\Delta q$ was achieved when $\Delta T$ was increasing, with a maximum $\Delta q$ at 234 kW/m$^2$ when $\Delta T$ was 16.8 K; thus, the $q_{SH}$ increased to more than 2 times that of $q_P$. However, when $\Delta T$ was more than 16.8 K, $\Delta q$ decreased with increasing $\Delta T$.

3.4.2. Heat transfer coefficient vs. temperature difference

The CHT coefficients $h_c$ were analyzed, as shown in Fig. 13. The $h_c$ of the superhydrophobic copper surface was distinctly higher than that of the pure copper surface. Here for convenience, the $h_c$ of the pure copper heat sink is denoted as $h_{CP}$. From the figure, one can see that the $h_{CP}$ was slightly changed within the measured $\Delta T$ range, and the trend of $h_{CP}$ was falling with increasing $\Delta T$, while the average $h_{DP}$ was approximately 12,900 W/m$^2$-K, which is comparable to Ma et al. [21,39].

The $h_c$ of the superhydrophobic condensation surface is denoted as $h_{CSH}$. Generally, the $h_{CSH}$ was higher than $h_{DP}$ at the same $\Delta T$ because the greater heat transfer flux of the superhydrophobic copper surface resulted from the shortened cycle time of droplet condensation, nucleation, and droplet falling. From the plot, the $h_{CSH}$ can be seen to be decreasing with increasing $\Delta T$, with no stable trend found. Compared with the results of other works [21,28,39], the $h_{CSH}$ in this study decreased more rapidly. Some possible reasons for this were already addressed in the previous section regarding the relationship between $q$ and $\Delta T$. However, another possible reason is found in Yang & Gu [19], who suggested that $h_c$ was also contributed to by the thermal resistance of the coating film. Therefore, this also suggests that the most likely reason for the steep decrease of $h_{CSH}$ is because of the thermal resistance resulting from the degradation of the EGC-1720 coating layer on the surface. This constitutes a vital issue for development and improvement in the future.

4. Conclusions

In this study, a developed method of surface modification was applied to successfully produce a superhydrophobic copper heat sink. For CHT study, the durability of two heat sinks was investigated, and these tests supplied a reference for the appropriate operation time period. Surface condensation features were used to confirm the results obtained by the durability test. Afterwards, CHT experiments were executed and yielded results superior to those of other studies that also used a monolithic copper heat sink.

Therefore, some contributions of this paper can be addressed as follows: first, a concise and low-cost surface modification method was able to be applied to a commercial heat sink to develop a superhydrophobic heat sink. The method could be a candidate for practical application in mass fabrication. Second, the durability of the CHT performance of the EGC-1720 coating layer was outlined, thereby providing a reference for future use. A shortcoming was the poor bonding between the EGC-1720 coating layer and copper oxide surface under vapor condensation; however, its convenience in use is a factor worth further study and development. Lastly, an acceptable CHT performance of the superhydrophobic copper surface was obtained, the performance of which is comparable to that of other research groups. In summary, the superhydrophobic copper heat sink in this study has the potential to be applied in practice if the stability of the superhydrophobicity can be maintained for an extended period of time.

The stability of superhydrophobicity is influenced by the degradation of the surface coating. The possible solutions include replacement with a new coating material or improvement of the current coating technique to enhance bonding with the copper surface, both of which are still under investigation. Moreover, a stable surface coating is certainly required to sustain the long-term operation performance of CHT.

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Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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<tbody>
<tr>
<td>$A$</td>
<td>effective condensation area (m$^2$)</td>
</tr>
<tr>
<td>$C_p$</td>
<td>constant-pressure specific heat of cooling water (4.18 J/g-$\degree$C)</td>
</tr>
<tr>
<td>$h_c$</td>
<td>condensation heat transfer coefficient (W/m$^2$-K)</td>
</tr>
<tr>
<td>$h_{CP}$</td>
<td>condensation heat transfer coefficient of pure copper surface (W/m$^2$-K)</td>
</tr>
<tr>
<td>$h_{CSH}$</td>
<td>condensation heat transfer coefficient of superhydrophobic copper surface (W/m$^2$-K)</td>
</tr>
</tbody>
</table>
\(k_{c}\) thermal conductivity of copper (401 W/m-K)
\(m\) mass flow rate of cooling water (g/s)
\(q\) heat flux (W/m²)
\(q_{p}\) heat flux of pure copper surface (W/m²)
\(q_{S\text{SH}}\) heat flux of superhydrophobic copper surface (W/m²)
\(T_{1}\) inlet temperature of cooling water (°C)
\(T_{2}\) outlet temperature of cooling water (°C)
\(T_{3}\) vapor temperature at vapor inlet (°C)
\(T_{4}\) inner temperature of heat sink (°C)
\(\Delta T_{\text{surf}}\) condensation surface temperature (°C)
\(\Delta q\) heat flux difference between superhydrophobic copper surface and pure copper surface (W/m²)
\(\Delta T\) temperature difference between \(T_{3}\) and \(T_{\text{surf}}\) (K)
\(\Delta x\) distance between Thermocouple 4 and the condensation surface (mm)

**Acronyms**

CHT condensation heat transfer
DWC dropwise condensation
FWC filmwise condensation
SAM self-assembly monolayer
NCG non-condensable gas
PC personal computer
SEM scanning electron microscope
WCA water contact angle
CAH contact angle hysteresis

**References**


