Condensation Heat Transfer Correlation for Micro/Nanostructure Properties of Surfaces
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ABSTRACT: Condensation, which can be observed in nature as a phase change heat transfer phenomenon, is a critical phenomenon in industrial fields such as power generation, water desalination, and environmental control. Many existing studies have applied surfaces with different wettability by controlling the surface topology to enhance condensation heat transfer. However, the industrial applicability is close to zero due to the limited size and shape of surfaces and low supersaturation conditions. Here, we regulate the surface topology of large-area copper tubes, which are representative industrial metals. We fabricated four copper tubes with different surface structures. We analyzed the condensation phenomenon of the modified tube under specific supersaturation conditions by measuring the overall heat transfer coefficient. We analyzed the condensation phenomenon by measuring the condensation heat transfer coefficient. We have recognized that there is a difference between the maximum droplet radius and the droplet detaching frequency depending on the size and shape of the structure. We measured the contact angle and contact angle hysteresis to accurately analyze the droplet behavior on each surface. As a result, we show that there is a correlation between contact angle hysteresis (CAH) and the total heat transfer coefficient, indicating heat transfer performance. These findings can be applied when evaluating surfaces with excellent condensation heat transfer performance for use in real industrial environments, which can dramatically reduce time and cost.

INTRODUCTION
Condensation heat transfer is attracting considerable interest in terms of energy efficiency since it can transfer a lot of heat even at a small temperature difference.1−4 Condensation has many applications in the field of thermal power plants,5,6 water desalination and harvesting,7,8 and electronics cooling.9,10

A common condensation phenomenon observed in metals is filmwise condensation. It means that the liquid condenses in the form of a film, which lowers the heat transfer efficiency of the surface.11,12 Meanwhile, if a surface is changed with the surface morphology and surface chemistry, the vapor condenses in the form of droplets. We call this phenomenon dropwise condensation. When a droplet reaches a certain size, it flows down the surface, fusing with other droplets. Due to this, the removal rate is faster than the formation rate of the droplet, so the heat transfer efficiency is very high. Existing papers have revealed that dropwise condensation shows overwhelmingly better heat transfer performance than filmwise condensation.13−20

For this reason, dropwise condensation studies have been actively conducted for several centuries, and many studies have induced dropwise condensation through surface property modification,21,22 altering the working conditions23 and changing the working fluid.24 While early studies implemented dropwise condensation by implementing a hydrophobic surface through the simple surface coating. Later studies
obtained a sustainable and high droplet removal efficiency by creating micro/nanosized structures on the surface.

Ghosh et al.25 implemented a hydrophilic—hydrophobic patterning surface that biomimics the zigzag-shaped surface of a banana leaf through laser processing and chemical etching. During the condensation experiment, dropwise condensation and filmwise condensation occurred at the same time, and the droplets generated from dropwise condensation were rapidly absorbed into the filmwise condensation area, greatly improving the heat transfer performance. They found that the maximum diameter of the droplets was strongly related to dropwise condensation.

Miljkovic et al.26 demonstrated through experiments that the heat transfer efficiency was higher on the nanostructured surface than on the previously studied polymer surface under the same conditions. Rongfu Wen et al.27 confirmed that the condensed droplets quickly disappear due to the difference in vapor density between the structures and the outside through the dense nanostructures. Through this, the research team dynamically analyzes and proves the mechanism by which condensation occurs in nanostructures. Jian Xie et al.28,29 conducted condensation experiments on the polymer surface and the nanostructured surface for one week and compared the dropwise condensation results of the two. In addition, the researchers theoretically explain the lowering of the heat transfer coefficient due to the deformation of the nanostructure.

Chehrghani et al.30 demonstrated that superhydrophobic nanostructures can be utilized to improve the condensation heat transfer performance in flow condensation. They observed flow condensation on vapor flows with different vapor qualities, pointing out that conventional condensation studies on superhydrophobic surfaces have been performed under ideal working conditions. They changed the values of steam mass fluxes and cooling water mass flow rate and investigated droplet dynamics such as droplet diameter distribution and droplet departure diameters through visualization studies.

Thomas and Mahapatra31 applied three hydrophobic coatings on an aluminum surface having micro/nanocomposite structures, conducted condensation experiments for each surface, and analyzed condensation phenomena. As a result, all three surfaces had the same contact angle hysteresis, but the condensation patterns were different depending on the coating agent. As a result of deriving the heat transfer coefficient for each surface, the heat transfer efficiency was rather low on the surface exhibiting the jumping droplet phenomenon, proving that the effect of sweeping is prominent on the surface exhibiting the dropwise condensation phenomenon.

However, recent studies related to dropwise condensation have common limitations. First, since previous studies observed the condensation pattern and performance only on one surface structure, the identification of the condensation pattern according to the size and shape of the structure was not made. Second, since the existing studies were performed on a laboratory scale of less than 100 mm, it may be difficult to apply them to actual industries. Third, it is difficult to predict the condensation pattern of a surface having a curvature by observing the condensation phenomenon on a flat plate.

In this study, we set shape, size, and single/hierarchical as the main parameters of surface structure related to condensation performance. First, we chose dendrite structures to control the shape of the structures equally and only to differ in size. The dendrite structures could be realized from nano to micro sizes depending on the process conditions. Second, to control the scale of the structure in the same way and to change only the shape, we fabricated cubic structures with microscale and compared the results with the microdendrite structures above. Finally, to confirm the difference between the microsingle structure and the micro/nanohierarchical structure, we implemented micro/nanocubic structures in which nanostructures are stacked on top of microcubic structures and compared the results. Through this, we identified the factors of the surface structure that have a major influence on the condensation performance and proved the optimal structure that shows the best condensation performance on a large-area surface with curvature. In addition, we conducted condensation experiments on a large-area surface under conditions similar to those of an industrial environment. In addition, we conducted a study with high potential for industrial application by conducting condensation experiments on a large-area surface under harsh conditions.

### RESULTS AND DISCUSSION

**Data Reduction.** The performance of each specimen was compared by the overall heat transfer coefficient ($U$) evaluated by the iterative method.39 The calculation process for the $U$ value is as follows. The heat rate of each specimen was calculated using a flow rate of a condensate, $m_{cond}$ (eq 1). Since it was impossible to implant a thermocouple on the surface of the specimen without damaging the structured surface, we assumed $T_{wet}$ to calculate the modified latent heat of water vapor (eq 2) and heat rate, $Q$ (eq 3). $h_g$ is the latent heat of saturated water vapor and $C_{p,fg}$ is an isobaric heat capacity. $R_{cond}$ is induced from the total thermal resistance of the system (eq 4). $R_{conv}$ is a reciprocal of convective heat transfer coefficient calculated using the Petukhov–Popov correlation. By varying

| Saturation pressure | Reynolds number 10,000 | Reynolds number 20,000 |
|---------------------|------------------------|------------------------|
| 0.2 bar             | Condition #1            | --                     |
| 0.4 bar             | Condition #2            | Condition #5           |
| 0.6 bar             | Condition #3            | Condition #4           |

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**Figure 1.** Condensation experiment conditions. (a) Major two variables of the experimental conditions. (b) Graph showing the increase in supersaturation by each condition.
the value of $T_{\text{surf}}$, the iteration was conducted until the error between $Q$ and $Q_\dot{}$ became less than 0.1%. Finally, we obtained the total heat transfer coefficient (eq 5).

$$Q = m_{\text{cond}}h^*$$  \hspace{1cm} (1)

$$h^* = h_{fg} + C_p(T_{\text{sat}} - T_{\text{surf}})$$  \hspace{1cm} (2)

$$Q = \frac{1}{R_{\text{cond}}}(T_{\text{sat}} - T_{\text{surf}})$$  \hspace{1cm} (3)

$$R_{\text{cond}} = \frac{1}{UA_{\text{surf}}} - R_{\text{conv}} - R_w = \frac{1}{h_{\text{cond}}A_{\text{surf}}}$$  \hspace{1cm} (4)

$$U = \frac{Q}{A_{\text{surf}}\Delta T_{\text{LMTD}}}$$  \hspace{1cm} (5)

Condensation Experiment Condition. For the condensation experiment on the superhydrophobic copper surface of various structures, the experimental conditions were set as shown in Figure 1. As a variable of this condensation experiment, two major parameters were set: the flow rate of the coolant and the saturation pressure inside the chamber. The flow rate of the coolant was set to the Reynolds number of 10,000 and 20,000 and the saturation pressure was set to 0.2, 0.4, and 0.6 bar. When the Reynolds number was set to 10,000, the experiment was carried out by increasing the saturation pressure from 0.2 to 0.6 bar, and Conditions 1 to 3 were

Figure 2. Superhydrophobic surface modification process of three-dimensional large-area copper tube: (a) nanodendrite (ND), (b) microdendrite (MD), (c) microcubic (MC), and (d) micro/nanocubic (MNC). Photograph courtesy of “Y.S.” Copyright 2022.

Figure 3. Surface modeling images and SEM images for each structure: (a) nanodendrite (ND), (b) microdendrite (MD), (c) microcubic (MC), and (d) micro/nanocubic (MNC). Photograph courtesy of “Y.S.” Copyright 2022.
The oxidation process was carried out with a 2.5 M sodium hydroxide solution and 0.2 M ammonium persulfate mixture solution at 4 °C for 30 min and then acid solution at room temperature for 5 min and then etched in a right-angle form different from the dendrite structure. In particular, in the case of the fourth micro/nanocubic structures, it can be seen that several layers of nanocubic structures are formed on the microcubic structures (Figure 3d). In particular, in the case of the fourth structure, it can be seen that the hierarchical structures are a mixture of micro- and nanostructures.

Fabrication Process of the Four Superhydrophobic Copper Tubes. We first fabricated nanodendrite and microdendrite surfaces to compare surfaces with the same structure but different scales. Next, microcubic surfaces were secondarily fabricated to compare surfaces with the same scale but different shapes. Finally, micro/nanocubic structures were fabricated to compare the differences between microsingle structures and micro/nanohierarchical structures.

The prepared copper tubes were immersed in 20% nitric acid solution at room temperature for 5 min and then ultrasonically cleaned in ethanol for 10 min to remove the existing oxide layer. To make a copper nanodendrite structure, the oxidation process was carried out with a 2.5 M sodium hydroxide solution and 0.2 M ammonium persulfate mixture solution at 4 °C for 30 min (Figure 2a). To make a copper microdendrite structure, the initial oxidation process was carried out with a 0.02 M potassium chlorate solution in a 60 °C oven for several hours. To form a stable oxide layer, a trace amount of sulfuric acid was added to keep the pH of the solution constant at 3.0. An additional oxidation process was conducted with the solution that was added with 0.1 M cupric sulfate pentahydrate for several hours. The green copper microdendrite structure was established on the surface (Figure 2b).

To form a copper micro/nanohierarchical structure, the oxidation process was first carried out with a mixture solution of a 1:3 ratio of 38% iron chloride solution and 35% hydrochloric acid solution at ambient temperature for several hours (30 h) (Figure 2c). Next, an additional oxidation process was carried out with ammonium hydroxide (NH₄OH) at 60 °C for several hours (45 h) (Figure 2d). After all oxidation, the tubes were rinsed with deionized (DI) water for 5 min in a sonication water bath and dried in an oven at 60 °C for 15 min. The tubes were chemically modified with 0.25 wt % n-hexane solution of 1H, 1H, 2H, 2H-perfluorodecyltrichlorosilane (FDTS) for 30 min and were dried in the oven at 60 °C for 15 min. After the process, a self-assembled monolayer was formed on the tubes.

Characterization of Four Superhydrophobic Copper Surfaces. To reveal the surface morphology of each surface, we performed field emission scanning electron microscopy (FE-SEM) analysis. Figure 3 shows FE-SEM images of four structured copper surfaces. First, it can be seen that the nanodendrite structures have a needle-like structure with the size of several nanometers formed on the surfaces. The needle-like structures are entangled with each other and have a similar shape to that of a dendrite (Figure 3a). Second, in the case of microdendrites, the shape is similar to the first structure, showing a shape in which the needle-like structure extends from one point. The difference is that the size is about a few micrometers (Figure 3b). Third, in the case of the microcubic structures, it can be seen that the microcubic structures are etched in a right-angle form different from the dendrite structures discussed above (Figure 3c). In the case of the micro/nanocubic structures, it can be seen that several layers of nanocubic structures are formed on the microcubic structures (Figure 3d). In particular, in the case of the fourth microstructure, it can be seen that the hierarchical structures are a mixture of micro- and nanostructures.

Condensation Heat Transfer Performance Evaluation. To evaluate the heat transfer performance of each specimen, we conducted a condensation experiment on a horizontal tube in the experiment. Figure 4 shows the U values calculated according to each condition in four horizontal tubes.

First, we confirmed that the nanostructured surface exhibited higher U values than the microstructure surface. Through these results, we could confirm that even if the structure had a similar shape, the heat transfer performance varied depending on the scale of the structure. We pointed to the difference in the aspect ratio of the structure, which was caused by the difference in distance between the structures, as the cause of this phenomenon. In the case of the nanostructure, the distance between the structures was smaller than that of the microstructure, so the aspect ratio appeared larger, and it was difficult for droplets to penetrate between the structures during the condensation process. In other words, in the process of coalescence after nucleation of droplets occurred, the suspended state, not the pinned state, was maintained.

We confirmed that in the suspended state, the separation of the droplets occurred faster, and thus the heat transfer efficiency increased. In addition, we confirmed that higher U values were measured in the microdendrite structure than in the microcubic structure. Through these results, we proved that even if the size of the surface structure was the same, dissimilar condensation occurred when the shape of the structure was different. In the case of the cubic structure, the aspect ratio was small and the maximum droplet radius was larger, so detachment did not occur well. In other words, the number of attached droplets was larger and this acted as a big factor that hindered heat transfer.

However, in the case of the micro/nanocubic structure in which the nanostructure was created on the microcubic structure, it can be seen that the hierarchical structures are a mixture of micro- and nanostructures. The prepared copper tubes were immersed in 20% nitric acid solution at room temperature for 5 min and then ultrasonically cleaned in ethanol for 10 min to remove the existing oxide layer. To make a copper nanodendrite structure, the oxidation process was carried out with a 2.5 M sodium hydroxide solution and 0.2 M ammonium persulfate mixture solution at 4 °C for 30 min (Figure 2a). To make a copper microdendrite structure, the initial oxidation process was carried out with a 0.02 M potassium chlorate solution in a 60 °C oven for several hours. To form a stable oxide layer, a trace amount of sulfuric acid was added to keep the pH of the solution constant at 3.0. An additional oxidation process was conducted with the solution that was added with 0.1 M cupric sulfate pentahydrate for several hours. The green copper microdendrite structure was established on the surface (Figure 2b).

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structure, the heat transfer performance was better than that in the microcubic structure as well as the microdendrite structure. The reason is that the number of valleys per single area increased due to the nanostructure, which decreased the maximum droplet radius and increased the detachment frequency. In summary, the nanodendrite structure with the highest number of structure valleys per area showed outstanding heat transfer efficiency. On the contrary, the microcubic structure with a small number of valleys and a small aspect ratio showed the lowest heat transfer coefficient.

Previous studies were conducted focusing on the dropwise condensation phenomenon in a limited situation with low supersaturation, that is, a small subcooling temperature. In addition, the size of the specimen is limited to a maximum of 50 mm and the shape is limited to a flat plate. There is a big gap in applying these research results to actual industrial heat exchangers. We conducted droplet analysis experiments to determine whether each surface is superhydrophobic and to see if there is a correlation between contact angle hysteresis (CAH) and heat transfer coefficient. The contact angle properties results of four different surfaces are shown in Table 1. We confirm that the apparent contact angle ($\theta_0$) is over 150° on all surfaces, indicating superhydrophobicity. We measure the receding angle ($\theta_r$) and advancing angle ($\theta_a$) on

| Surface          | $\theta_0$ (deg) | $\theta_a$ (deg) | $\theta_r$ (deg) | CAH (deg) | Wetting Behavior |
|------------------|------------------|------------------|------------------|-----------|-----------------|
| bare             | 68 ± 2           | 76 ± 5           | 48 ± 4           | 28        | wetted          |
| nanodendrite     | 162 ± 4          | 166 ± 5          | 152 ± 5          | 14        | suspended       |
| microdendrite    | 154 ± 2          | 159 ± 6          | 121 ± 4          | 38        | partial wetted  |
| microcubic       | 152 ± 3          | 158 ± 6          | 105 ± 8          | 52        | partial wetted  |
| micro/nanocubic  | 158 ± 3          | 163 ± 6          | 141 ± 2          | 22        | suspended       |

Figure 5. Results of durability evaluation for each structure with conditions 1–5. The overall heat transfer coefficients at the bare and four different structured surfaces are shown: (a) nanodendrite (ND), (b) microdendrite (MD), (c) microcubic (MC), and (d) micro/nanocubic (MNC).
all surfaces and calculate CAH from the difference between the two \((\text{CAH} = \theta_{\text{r}} - \theta_{\text{a}})\). In the case of CAH, the difference was remarkable. CAH increases gradually in the order of nanodendrite, micro/nanocubic, microdendrite, and microcubic.

We note that this tendency of CAH was surprisingly equal to that of the \(U\) value. We confirmed that the smaller the CAH value, the larger the \(U\) value. We judged that the reason for the correlation between the two values was that the frequency of droplet removal and CAH were directly related. The small CAH meant that the droplet exists in a suspended state rather than a pinned state, so when tilting, it slid down easily by gravity. Conversely, the fact that the CAH was large meant that when tilting was in progress, the droplet was in a state that was disturbed by the structure, that is, it existed in a pinned state. As the supersaturation increased during condensation, the state tendency of the coalescence droplets generated was a similar mechanism. Therefore, we evaluated the state tendency of condensed droplets to be predictable through CAH measurement. Also, since there was a close relationship between the droplet state tendency and the droplet detaching frequency, we considered it to be sufficient to predict the heat exchange performance.

We retested the superhydrophobic copper tubes by setting 1 week and long-term (more than 3 months) conditions, respectively, to check whether the condensation performance deteriorated over time. In the case of the long term, we attempted to set the same period for each structure, but there was a slight difference in the period due to the influence of the optimization of the experimental equipment and the schedule (nanodendrite: 115 days, microdendrite: 108 days, microcubic: 96 days, micro/nanocubic: 92 days). The result is shown in Figure 4. Overall, there was no significant difference in the condensation performance over time. However, in the case of microcubic, it can be seen that the performance becomes similar to that of bare after a long time has passed. It was analyzed that the physical properties of the existing structure were lost due to the high degree of oxidation in air. On the other hand, in the case of nanodendrite, the condensation performance was maintained well even after a long-term. Through this, we confirmed that the above surfaces did not have any problems with repeated condensation tests and long-term use (Figure 5).

## CONCLUSIONS

We evaluated that the existing studies are focused on the laboratory scale, so there are significant limitations to be applied to the actual industry. No matter how high the heat transfer efficiency is measured, if it is only a small part, it is not certain whether the heat transfer efficiency increases when applied to an actual heat exchanger. In addition, the nanostructured surfaces used in previous studies are not currently applicable to industrial heat exchanger surfaces, since the process conditions are complex and cost is high. We have overcome these limitations through the following studies.

1. We modified the surface of 500 mm long copper steam condenser tubes using a scalable micro/nanofabrication method with simple process conditions and low cost. The process conditions consisted of cleaning/etching, oxidation, and self-assembly monolayer (SAM) coating, and the following four structures were implemented: nanodendrite (ND), microdendrite (MD), microcubic (MC), and micro/nanocubic (MNC).

2. We performed SEM analysis to confirm whether the surface structure was stably formed. In addition, we confirmed that all surfaces had the same superhydrophobic properties (apparent contact angle >150) through contact angle measurement. However, we confirmed that the CAH values were different because there were differences in the size and shape of each structure.

3. We evaluated the total heat transfer coefficient and heat resistance through a vapor condensation experiment conducted with a bare copper tube and four copper tubes in the same experimental apparatus. We observed how the condensation behavior changed according to each supersaturation condition and evaluated the average \(U\) value for each surface.

4. We investigated how the change in the condensation behavior varies according to the CAH value of each surface and derived the correlation with the average \(U\) value. Through the derived correlation, we confirmed that the smaller the CAH, the more the dropwise condensation was maintained even at the high supersaturation, and the \(U\) value was higher. With these findings, we proved that CAH was a major cause of direct influence on condensation heat transfer even on large-area surfaces. Our findings could predict the heat transfer efficiency of large-area surfaces using only CAH measurements. Since these research results could be applied to the industrial field, it would have a great influence on related studies. As a future study, we emphasized the surface fabrication process and condensation experiments that could be applied to large-area heat exchangers and show low CAH. If the condensation phenomenon is proven on multiple surfaces, it would be possible to drastically reduce the time and cost incurred when searching for an excellent surface structure applicable to the surface of an industrial heat exchanger.

## EXPERIMENTAL SECTION

### Materials

All copper tubes used in the condensation experiment have the following specifications: an outer diameter of 25 mm, a thickness of 2 mm, and a total length of 500 mm. Nitric acid solution \((\text{HNO}_3)\), ethanol \((\text{C}_2\text{H}_5\text{OH})\), potassium chlorate \((\text{KClO}_3)\), sulfuric acid \((\text{H}_2\text{SO}_4)\), cupric sulfate pentahydrate \((\text{CuSO}_4\cdot\text{H}_2\text{O})\), sodium hydroxide \((\text{NaOH})\), ammonium persulfate \((\text{NH}_4)\text{S}_2\text{O}_8)\), iron chloride \((\text{FeCl}_3)\), hydrochloric acid \((\text{HCl})\), ammonium hydroxide \((\text{NH}_3\cdot\text{H}_2\text{O})\), and \(n\)-hexane \((\text{CH}_3\cdot\text{CH}_2\cdot\text{CH}_2\cdot\text{CH}_2\cdot\text{CH})\) were purchased from SAMCH-UN Chemical, Korea. \(1\text{H},1\text{H},2\text{H},2\text{H}-\text{Perfluorodecylnitrilchlorosilane} (\text{FDTS})\) was purchased from Alfa Aesar, AR.

### Characterization

The bare and four structured copper surfaces were investigated through a scanning electron microscope (JEOL Ltd., Japan). We measured the contact angle properties utilizing a droplet analyzer (Smart Drop, FemtoFAB, Korea). The contact angle of each specimen was measured by specifying five different points for each surface, and the mean and standard deviation at five points were calculated. The contact angle hysteresis (CAH) was measured through the tilting plate method and by slowly tilting the cradle at an angle between 0 and 90°.

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Notes
The authors declare no competing financial interest.

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NOMENCLATURE

| Symbol | Description | Unit |
|--------|-------------|------|
| Q      | heat transfer rate | [W]  |
| m      | mass flow rate    | [kg/s] |
| hfg    | latent heat      | [J/kg] |
| hfg*   | modified latent heat | [J/kg] |
| C_p    | isobaric heat capacity | [J/kg K] |
| T      | temperature       | [°C] |
| U      | overall heat transfer coefficient | [W/m² K] |
| LMTD   | logarithmic mean temperature difference cond | condensation |
| sat    | saturation surf   | surface |
| f      | film w            | wall |
| A      | area [m²] R       | thermal resistance [K/W] |
| h      | heat transfer coefficient | [W/m² K] |

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