A Study of Droplet-Behavior Transition on Superhydrophobic Surfaces for Efficiency Enhancement of Condensation Heat Transfer

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ABSTRACT: Enhancement in heat-transfer performance via dropwise condensation on superhydrophobic surfaces is much greater than that realized via generic condensation on a regular surface. However, if the supersaturation level during condensation increases above a specific value, water may seep to greater depths between structures. This may lead to attached condensation, which reduces condensation heat-transfer efficiency below that of ordinary surfaces. Therefore, it is critical to avoid the occurrence of supersaturation when superhydrophobic surfaces are employed in condenser design. The proposed study presents a simple method for regulating supersaturation on the laboratory scale. Experiments concerning droplet behavior on a superhydrophobic plate were performed to investigate droplet detachment and attachment in accordance with the surface and droplet temperatures. Results obtained have been represented as a "droplet-behavior map", which clearly depicts boundaries dividing the detachment and attachment regions. The supersaturation threshold obtained from the said map has been compared against results obtained from condensation heat-transfer experiments performed in an actual condenser environment. As observed, the two results demonstrate excellent agreement. Although superhydrophobicity of surfaces remains unchanged at room temperature, changes may occur in the extent of the supersaturation section, which improves condensation heat-transfer performance, depending on the surface-structure complexity. Therefore, droplet-behavior mapping has been used in this study to determine the available supersaturation section in accordance with the variation in surface roughness. Results confirm that the available supersaturation region increases with increasing surface roughness and structural complexity. Therefore, prior to applying superhydrophobicity to condensers, droplet-behavior mapping must be performed to avoid operation under the supersaturation conditions, which causes attached condensation.

1. INTRODUCTION

Generally, the heat-transfer efficiency of condensers has a considerable effect on the energy efficiency of power plants. Energy efficiency has assumed increased importance in recent years owing to the limited availability of conventional energy sources, and the same is true of condenser efficiency as well. Condensation heat transfer represents the core condenser phenomenon involving cooling of heated vapor produced after turbine operation. Condensers are not only used in such plants as petrochemical, thermoelectric power, and nuclear power but also in vehicles and electric devices. Consequently, several studies have been performed to improve condensation heat-transfer performance and, hence, enhance condenser efficiency. Generally, dropwise condensation is known to result in heat-transfer efficiency values several times higher than those realized via film condensation.5–6 Dropwise condensation can be realized by inducing hydrophobicity on condenser surfaces. Applications involving use of hydrophobic surfaces with contact angles (CAs) exceeding 150°—called superhydrophobic surfaces—have been reported to result in realization of effective dropwise condensation.7 Superhydrophobicity can be realized by performing micro-nanostructure fabrication on condenser surfaces followed by self-assembled monolayer (SAM) coating of a hydrophobic polymer.8–14 However, some extant studies report that the condensation heat-transfer
efficiency may drop significantly despite use of a super-
hydrophobic surface. Several researchers have attempted
to investigate reasons behind this performance degrada-
tion. Miljkovic et al. reported that there exist four types
of condensation—jumping, dropwise, flooded, and filmwise—
in the order of efficiency. Jumping condensation is observed at
low supersaturation levels ($S = \frac{P}{P_{sat}}$, $S = 1.08$), and an increase
in the supersaturation level results in transition to flooded
condensation. Ji et al. fabricated a superhydrophobic surface
on a condenser tube and analyzed the condensation
phenomenon. Through their experiments, it was realized that
the condensation heat-transfer performance of superhydro-
phobic surfaces may be lower than that of regular surfaces
under certain operating conditions. This reduction in heat-
transfer performance can be explained based on the super-
saturation concept. Enhanced condensation heat-transfer
phenomena, such as jumping or dropwise condensation, are
usually observed at low supersaturation levels. Although the
efficiency of flooded condensation is lower than that of
dropwise condensation, it is still higher than that of a regular
surface with film condensation. However, when the super-
saturation level exceeds a certain level, the condensation heat-
transfer efficiency dramatically declines owing to moisture
infiltration into gaps within the surface nanostructure. This
phenomenon is referred to as attached condensation and
causes the condensation heat-transfer efficiency to drop below
that offered by regular film condensation. Superhydrophobic
surfaces capable of preventing the occurrence of attached
condensation are difficult to fabricate. However, high
condensation heat transfer can be maintained by avoiding
the occurrence of the condition that causes attached
condensation. Jo et al. investigated factors that cause a decline
in condensation performance of superhydrophobic surfaces.
In their report, the critical gap size of the surface nanostructure
was used as a criterion of water infiltration defined as a
function of supersaturation. The report states that surface
nanostructures with a sufficiently small gap size help prevent
droplet penetration during condensation, thereby preventing
performance degradation due to attached condensation.

Droplet-behavior (DB) experiments were performed in this
study to analyze the condensation phenomenon observed
during condensation heat transfer. In addition, a method to
determine regions that maintain improved condensation
performance by changing the critical gap size or adjusting
supersaturation levels has been developed. A droplet-behavior
map has been derived by conducting experiments involving use
of a superhydrophobic plate, and the said map was evaluated
by performing a condensation experiment involving use of a
tube with an identical superhydrophobic surface under an
actual condenser environment. Subsequently, conditions for
surface-structure selection and operation favorable for
enhanced condensation heat-transfer performance were
derived by performing droplet-behavior experiments. Findings
of this study are expected to dramatically expand the scope of
industrial applications of heat-transfer enhancement technol-
ogies.

2. RESULTS AND DISCUSSION

2.1. Fabrication and DB Mapping of Superhydropho-
bic Aluminum. Figure 1a depicts changes in the surface
wettability of aluminum during treatment. Etching in HCl
solution results in formation of a cube-like microstructure
on aluminum plates, whereas deionized (DI) water oxidation
causes formation of a flake-like nanostructure. Thus,
macroscopic micro/nanostructures are formed on aluminum
plates subjected to these processes (Figure 1b). Application of
the hydrophobic polymer ($1H,1H,2H,2H$-perfluorodecyltri-
chlorosilane, PFOTS) SAM coating changes the nature of
the surface from superhydrophilic to superhydrophobic
without damaging its internal structure. The phenomenon of
superhydrophilicity can be explained using the well-known
Cassie–Baxter theory, and hydrophobicity of a surface can be
significantly enhanced by increasing its roughness.

The temperature difference between the surface and water
droplets was a dominant parameter on the phenomenon of
droplet attachment, while the droplet behavior was unaffected
by the change of surface inclination. Therefore, a surface
inclination of 70° was decided as the best angle to see the
attachment and detachment of droplets. Figure 1c,d depicts
instances from videos (recorded using a high-speed camera)
capturing the sequence of events from the moment water
droplets contact the surface until after they separate from the
pipette (Videos S1, S2). Both experiments were performed on
one surface of the samples. The droplet temperature ($T_d$) was
toggled between room temperature ($T_w$) and 85 °C, whereas
the surface temperature ($T_s$) was maintained at $T_s$. It is difficult

Figure 1. (a) Water contact angle change by the surface treatment process. (b) SEM images of the fabricated M-N-F aluminum surface. Photos of the droplet behavior using a high-speed camera in the (c) detachment condition ($T_d = T_w$, $T_s = T_i$) and (d) attachment condition ($T_d = 85^\circ C$, $T_s = T_i$), where $T_d$ and $T_s$ are the temperatures of the droplet and substrate, respectively.
for water droplets to remain attached to superhydrophobic aluminum surfaces maintained at room temperature even at very small inclination angles. Thus, on a surface inclined at 70°, water droplets always fall off neatly, as shown in Video S1 and Figure 1c. However, when hot water droplets having a large temperature difference with respect to the sample surface come in contact with the surface, they adhere to it (Video S2 and Figure 1d). This phenomenon occurs owing to the existence of a high supersaturation level $S$ at the interface between the droplet and surface due to the difference between saturated vapor pressure $P_{\text{wall}}$ at surface temperature and vapor pressure $P_v$ on the droplet surface. The supersaturation level $S$ was suggested in Miljkovic’s report as a variable for distinguishing condensation phenomena and can be defined as the ratio of vapor pressure to saturated vapor pressure corresponding to the surface temperature. \(^{16}\)

$$S = \frac{P_v}{P_{\text{wall}}} \quad (1)$$

Droplets tend to remain attached to superhydrophobic surfaces because water penetrates into nanoscale surface structures owing to its high vapor pressure. Figure 2 depicts the droplet-behavior map for attachment and detachment of droplets with surface and droplet temperatures varying between 30 and 100 °C, as obtained for the superhydrophobic aluminum samples fabricated in this study.

Figure 2a depicts results pertaining to droplets that remain attached to the micro-nano-fluorinated (M-N-F) superhydrophobic aluminum sample #1. As described in Figure 2a, the region of droplet attachment/detachment in accordance with the surface and droplet temperatures is defined as the droplet-behavior (DB) map. The DB map confirms that droplet attachment occurs under the condition of low surface and high droplet temperatures. DB mapping was performed for the three superhydrophobic aluminum samples fabricated using the same method. Results demonstrate formation of similar boundary lines for all three specimens (Figure 2b). Furthermore, the durability result of aluminum samples is depicted in Figure 2c, which confirms that no performance deterioration occurred after samples were immersed in water maintained at room temperature for 56 days. This suggests that uniform surface performance and excellent durability can be realized when the method for fabrication of superhydrophobic aluminum surfaces is applied to surfaces of condensation heat exchangers. To consider the effect of the surface structure, micro-fluorinated (M-F) aluminum surfaces were additionally fabricated, and corresponding results obtained have been compared in Figure 2b. Contact angles equaled 158.5 ± 1.5 and 168.9 ± 3.8° for the M-F and M-N-F aluminum surfaces, respectively; i.e., CAs of both surfaces exceed 150°. Thus, both surfaces can be considered superhydrophobic. However, as described in the DB mapping result, the boundary line corresponding to the M-F surface lies below that corresponding to the M-N-F surface. This phenomenon is discussed in Section 2.2.

The average supersaturation level $S_b$ of the boundary lines in the DB map corresponding to abovementioned aluminum surfaces was determined to be approximately equal to 8.7. 

Figure 2. (a) DB map of the Al M-N-F surface sample #1. (b) Boundary lines of Al M-N-F samples #1 to #3 and Al M-F samples #1 and #2. (c) Retrial of Al M-N-F after dipping in water at $T_r$ for 56 days.

Figure 3. (a) Area division by the boundary line of the DB map and $S_b$. $S_b$ of Al M-N-F is 8.7, and $S_b$ is 1. (b) Integrated experimental results of condensation experiments and droplet-behavior experiment.
Figure 3a depicts the DB map divided into three regions based on boundary lines corresponding to supersaturation values of 1 and 8.7. Condensation does not occur in region C since this is only possible in an environment where supersaturation exceeds 1 (S > S_c, (S_c = 1)). Additionally, it can be observed that dropwise condensation occurs in region B, whereas attached condensation occurs in region A, where S > S_c. The occurrence of other condensation phenomena, such as jumping, dropwise, and flooded condensations, cannot be easily observed in region B, and other additional variables are required to identify these.

The meaning of the supersaturation value of the boundary line S_c = 8.7 can be interpreted by comparing it against the results reported by Ji et al. in 2019.22 They analyzed the condensation phenomenon occurring in an actual condenser environment by using a superhydrophobic aluminum tube fabricated using the same method as that employed in this study for fabrication of superhydrophobic aluminum samples. Figure 3b depicts the graph indicating—on the DB map—the results obtained by performing the condensation heat-transfer experiment involving aluminum tubes. The average boundary line for samples #1, #2, and #3 is depicted, and results of the condensation heat-transfer experiment are marked separately by the condition number (Table S1). The experiment reported by Ji et al. was performed for condition numbers defined based on the Reynolds number and saturation pressure of coolants in an M-N-F aluminum tube. Dropwise condensation was observed under conditions #1 and #2 demonstrating performance improvements of the order of 105% on average, in terms of the total heat-transfer coefficient, compared to the bare tube. When flooded condensation was observed under conditions #3 and #4, the performance was observed to have improved by 16% on average compared to that of the bare tube. Attached condensation was observed under condition #5, where the heat-transfer performance was observed to decrease by 20% compared to that of the bare tube.22 In addition, two reproduction tests were performed as tests #6 and #7. Test #6 was performed to validate the total heat-transfer-coefficient result obtained under condition #2 after attached condensation was observed, whereas test #7 was performed to validate the total heat-transfer coefficient under condition #2 after flooded condensation was observed. Results obtained for tests #6 and #7 demonstrate noticeable degradation in performance in terms of the total heat-transfer coefficient post occurrence of attached condensation.22 What is especially noticeable in this result is that the attached-condensation condition of the tube experiment closely matches the attached region observed on the DB map. This suggests that the occurrence of attached condensation can be predicted using DB mapping.

In all the results of the above-described tests #6 and #7, the surface temperature can be observed to drop sharply and move toward the diamond mark. Test #6 results in lower surface temperature than test #7. Similar to the analysis result of the total heat-transfer coefficient, the supersaturation level in test #6 was observed to closely approach 8.7, which corresponds to the attached-droplet region on the DB map, after attached condensation was observed. Thus, it can be concluded that the closer the saturation level is to the attached-droplet region on the DB map, the lower is the observed condensation performance. The above analysis again demonstrates that the occurrence of attached condensation can be predicted via DB mapping, and the same can be quantified in terms of the supersaturation variable S.

### 2.2. Relationship between the Surface Structure and DB Map

Figure 2b depicts boundary lines observed on DB maps of the M-N-F and M-F aluminum plates. As can be observed, the region of attached droplets on the DB map for the M-F plate is larger than that corresponding to the M-N-F plate. A large region of attached droplets indicates poor condensation heat-transfer performance. This difference can be explained in relation to the structure size of each surface. In the SEM image of the M-F plate surface (Figure S1), a microstructure with a gap of 1.14 ± 0.13 μm was identified. On the other hand, while the M-N-F plate surface has a microstructure (1.05 ± 0.11 μm) of similar size, a nanostructure at the level of 44 ± 5 nm is hierarchically formed on the surface. Theoretically, the condensation condition under which droplets adhere to the surface owing to high supersaturation levels is affected by the size of gaps within surface structures, and this can be explained using the following equation.

\[ 2\sigma_l \cos \theta = -RT/v_l \ln\left(\frac{P_v}{P_{wall}}\right)x_{crit} \]  

(2)

Here, \( \sigma_l \) and \( \theta \) denote the interfacial force of liquid/vapor and intrinsic contact angle, respectively, whereas \( R, T, v_l, x \) and \( x \) denote the ideal gas constant, temperature, liquid molar volume, and size of gaps between nanostructures. The term \( x_{crit} \) denotes the critical gap size that prevents the occurrence of wet condensation at specific supersaturation values. Although it is difficult to calculate the actual value of \( x_{crit} \) owing to it being a theoretical variable, an important fact can be derived from the above equation. When a surface is hydrophilic (\( \theta < 90^\circ \)), the terms on the left of the equality sign in the above equation yield a positive number, and wet condensation occurs unconditionally regardless of the gap size in the section where the supersaturation level exceeds 1. However, if a surface is hydrophobic (\( \theta > 90^\circ \)), the smaller the gap size, the larger is the range of supersaturation levels causing dry condensation.

The critical value of supersaturation that causes the occurrence of attached condensation could not be determined from this result. However, improved heat-transfer performance can be obtained during operation under any oversaturated-vapor environment if seawater at room temperature is used within the condenser tubes of an actual power plant with the temperature of steam in contact with the tube being maintained between 30 and 80 °C. It is expected that realization of enhanced condensation performance of condenser tubes via use of the proposed technique would help us get one step closer to creating economic benefits through practical industrial application.

### 3. CONCLUSIONS

This study, through use of a simple experiment, proposes a method to determine operating conditions under which heat-transfer performance of condensers drops significantly. In this study, superhydrophobic surfaces were fabricated, and a droplet-behavior map, which clearly defines regions of droplet attachment and detachment divided by a boundary line, has been plotted. The supersaturation threshold that enables dropwise condensation was calculated considering the boundary line dividing the attachment and detachment regions. Additionally, implications of the said boundary line were analyzed by comparing experimental results against those obtained in an actual condenser environment. The reason
Figure 4. Experimental setup of droplet behavior. (a) The temperatures of both water and substrate are controlled by a hot plate and monitored by combined thermocouples. (b) Detail view of the droplet contact.

behind reduction in heat-transfer performance under conditions wherein droplet attachment occurs has been identified. In addition, the effect of the surface structure on the enhancement of heat-transfer performance was analyzed using aluminum samples. As observed, improvement in condensation heat-transfer performance is greater when the condensation surface is characterized by high roughness and high structural complexity. In conclusion, even when superhydrophobic heat pipes are used to facilitate droppwise condensation, performance deterioration may still be expected if attached condensation occurs within parts of the condenser. Furthermore, performance improvement, at sections where it can be achieved, is a strong function of surface roughness. Therefore, it is crucial to identify regions wherein attached condensation is likely to occur via use of DB mapping and, hence, avoid condenser operation in these regions.

4. MATERIALS AND METHODS

4.1. Surface Preparation and Characterization. Aluminum (Al5052; aluminum 97% (minimum)) plates measuring of 50 × 50 × 1 mm were used as substrates. Hydrochloric acid, sodium hydroxide, and n-hexane (SAMCHUN Chemicals, Republic of Korea) and PFOTS (Alfa Aesar, USA) were purchased from the market. All aluminum plates were first sonicated in ethanol and DI water. Precleaned aluminum plates were immersed in 1 M concentration NaOH solution at \( T_s \) for 1 min before subsequent washing in DI water. The aluminum plates were then immediately etched in a 3 M concentration HCl solution at \( T_s \) for 5 min and washed using DI water. Thereafter, the plates were oxidized in DI water at over 90 °C for 5 min and dried for 20 min in an oven maintained at 60 °C. For realization of hydrophobicity, fully dried aluminum plates were fluorinated using a 1000:1 mixture (by volume) of \( n \)-hexane and PFOTS for 10 min prior to being dried for over 20 min in an oven maintained at 60 °C.

Fabricated surfaces were closely observed using high-resolution field-emission scanning electron microscopy (SEM; JEOL; Japan). To evaluate the water repellency of fabricated surfaces, contact angles were calculated as the average of five measurements performed at different spots using an analyzer (Smart Drop; FemtoFAB; Korea).

4.2. Droplet-Behavior Experiment. Droplet-behavior experiments were performed in this study to investigate whether droplets remain attached or detached from the plate surface in accordance with changes in temperatures of the plate and water droplets falling on it. Figure 4 depicts the experimental setup used in this study. The hot plate on the left was used to control droplet temperature, while that on the right controlled the sample plate’s surface temperature. Superhydrophobic plate samples were fixed to the hot plate of the right using aluminum foil tape. To facilitate observation of the droplet-detachment phenomenon, the hot plate on the right was positioned with an inclination of 70° with respect to the horizontal direction. Surface temperatures of superhydrophobic plate samples were measured using K-type thermocouples with a precision of ±0.5 °C fixed to the right hot plate using aluminum tape.

Droplet temperature—i.e., temperature of DI water maintained in a beaker—was controlled by the left hot plate. Liquid temperature inside the beaker was measured using a K-type thermocouple. A micropipette with 3% accuracy was used to place 10 μL droplets on the surface of the plate sample. The water droplet hanging from the end of the pipette tip was placed on the surface for about 0.25 s, and then the pipette and water droplet were separated. Using multiple identical surfaces and repeated experiments showed that the results are the same regardless of the speed and direction of the droplets unless the speed is fast enough to free fall from a high place.

The droplet-behavior experiment can be described as follows. The experiment was performed to investigate whether droplets remain attached to the surface when the droplet temperature increases while the surface temperature remains fixed. Surface and droplet temperatures were adjusted in the range of 30–100 °C with 5 °C intervals (Figure 4b). When droplets remained attached to the surface, the result was displayed as O, whereas instances of droplet detachment were displayed as X. The experiment was repeated five times or more to ensure reliability of results obtained. A graph indicating regions of droplet attachment and detachment—marked by O and X, respectively—corresponding to changes in the surface and droplet temperatures was plotted and referred to as the droplet-behavior (DB) map.

The above experiment was performed using fabricated aluminum samples to understand the formation of the DB map. In addition, repeatability of the fabrication process was verified by performing the above experiment using multiple samples fabricated under identical conditions. Finally, the durability of fabricated samples was verified by performing the above experiment after immersing samples in water maintained at room temperature followed by drying.
ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.0c03081.

Recording of a droplet sliding on a superhydrophobic surface using a high-speed camera (Video S1) (MP4)

Recording of a droplet attachment on a superhydrophobic surface using a high-speed camera (Video S2) (MP4)

SEM images of the Al M-F surface. A micro-cubic-like structure is formed by HCl treatment (Figure S1) (PDF)

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Notes
The authors declare no competing financial interest.

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