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Realization of coalescence-induced condensate droplet jumping on a hierarchical porous superhydrophobic surface over a wide range of subcooling up to 20 K

Lin-Wei Mou, Tian-Yu Zhang, Jia-Yi Zhang, Jia-Qi Li, and Li-Wu Fan

AFFILIATIONS
1 Institute of Thermal Science and Power Systems, School of Energy Engineering, Zhejiang University, Hangzhou, Zhejiang 310027, People’s Republic of China
2 Department of Mechanical Science and Engineering, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801, United States of America
3 State Key Laboratory of Clean Energy Utilization, Zhejiang University, Hangzhou, Zhejiang 310027, People’s Republic of China

ABSTRACT
Coalescence-induced jumping of condensate droplets has been studied as an emerging mode for enhanced condensation heat transfer. However, the spontaneous droplet jumping phenomenon usually disappears at relatively higher degrees of subcooling due to the increase of nucleation density of condensate droplets. In this Letter, we show that self-propelled droplet jumping condensation can be realized surprisingly on a hierarchical porous superhydrophobic surface, having pores of the size of ∼20 µm with nanoscale dendritic structures, which was fabricated via a facile electrochemical deposition method. Droplet jumping condensation was always able to be sustained on this surface, making it never flooded even at elevated subcooling up to ∼20 K, although the intensity of droplet jumping degraded gradually with increasing the subcooling. It was hypothesized that the presence of microscale ridges among the pores, where nucleating droplets tended to appear and grow over the nano-structures on the ridge tips, serve as spatial separation barriers that prevent the surfaces from nucleation-density-modulated flooding at relatively high degrees of subcooling. As compared to conventional dropwise condensation on a smooth hydrophobic surface, significant heat transfer enhancement was able to attain for subcooling up to ∼7 K. The fabrication of this hierarchical porous superhydrophobic surface was deemed to be rapid, scalable, and highly-adaptive for curved surfaces on metallic bulk materials, e.g., copper tubes, with a great potential for industrial condensation applications.

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Condensation of vapor is an important phase change heat transfer process involved in a variety of industrial applications, such as desalination, power generation, refrigeration, and thermal management. A typical phenomenon observed in industrial condensation devices is the formation of a thin film of condensate over the cooling surface. Such filmwise condensation exhibits low heat transfer rate due to the added conductive thermal resistance by the liquid film that separates the vapor and the cooling surface. In contrast, improved heat transfer coefficient can be achieved by turning the filmwise condensation into the mode of dropwise condensation on surfaces with lower surface energy (or lower wettability as indicated by a greater apparent contact angle), where the condensate tends to form discrete droplets. The development of engineered hydrophobic/superhydrophobic surfaces with specially-designed micro-/nano-structures or a low-surface-energy coating have thus been sought for maintaining stable dropwise condensation.

During the past decade, an emerging mode of condensation has attracted increasing attention, i.e., self-repellent condensate droplets by coalescence-induced jumping on superhydrophobic surfaces.
Such spontaneous removal of micro-sized droplets off the cooling surface, due to the release of excess surface energy upon coalescence of droplets, is able to further enhance condensation heat transfer, as compared to the conventional droppwise condensation mode where removal of the droplets can only happen spontaneously until they grow to at least millimeter-sized under gravity. Recently, efforts have been dedicated to understanding and optimization of the droplet-jumping condensation process for enhanced heat transfer. In order to facilitate the release of excess surface energy over the threshold for droplet jumping, the adhesion and viscous dissipation of the cooling surface should be minimized. Hence, keen-edged and serried nano-structures are preferred to minimize the interface adhesion, which is influenced by the solid-liquid contact area, as well as to prevent penetration of condensates and vapor molecules into the nano-structures, which would be beneficial to the formation of Cassie-state droplets instead of the pinned Wenzel-state droplets during the initial stage of droplet growth. Moreover, as the diameter of droplets that are mostly possible to jump after coalescence lies in the range of 20−50 µm, specific micro-structures are also of importance for improving the frequency of droplet removal through the spatial confinement effect on the formed droplets.

Although the above-mentioned studies have gained insights into better manipulation of condensate droplet jumping on superhydrophobic surfaces, a main issue that limits the implementation of this highly-efficient mode of condensation heat transfer has been identified that droplet jumping can only be realized at relatively low degrees of subcooling (<∼5 K). As derived by Maa, the critical radius \( r_c \) of nucleating condensate droplets that can grow is given by

\[
r_c = \frac{2\sigma T_v}{\rho h \Delta T},
\]

where \( \sigma \) is the liquid-vapor surface tension, \( T_v \) the temperature of saturated vapor, \( \rho \) the density of condensate, \( h \) the latent heat of vaporization, and \( \Delta T \) the degree of surface subcooling (temperature difference between the saturated vapor and the cooling surface). It is obvious that with increasing the subcooling, the initial size of the nucleating droplets becomes smaller and the nucleation density increases, finally leading to flooding on the superhydrophobic surfaces when the distance between nucleating droplets reduces to become comparable to the characteristic length scale of the micro-/nano-structures.

A recent work by Wen et al. has demonstrated, for the first time, the achievement of stable droplet-jumping condensation over a wide range of subcooling up to 28 K, on a superhydrophobic surface coated with three-dimensional copper nanowire networks. The secret lies in the formation of interconnections among nanowires that improve significantly the mobility of droplets to prevent flooding at high degrees of subcooling. The straight copper nanowire arrays were grown by electrochemical deposition on a porous anodic aluminum oxide template, which was fabricated intentionally from an aluminum foil with ∼0.5% impurity. However, despite the presence of nanoscale bumps on the vertical walls of the nanowires, leading to the formation of interconnections among nanowires, the large length-to-diameter ratio (∼100) of these nanowires may cause them to bend and become lodging. Also, the electrochemical deposition process for the relatively long nanowires (20−30 µm) seems to be time-consuming, which took several hours (150−240 min).
As inspired by the idea of spatially suppressing the increase of nucleation density, here we report on an alternative approach to achieving stable droplet-jumping condensation. Although it was suggested that a surface with sub-micrometer-sized pores, which can avoid penetration of moisture and are less adhesive to micro-sized droplets, is in favor of realizing coalescence-induced jumping of condensate droplets, we demonstrate that the use of a hierarchical porous superhydrophobic surface, having a nominal pore size of ∼20 μm with nanoscale dendritic structures (see Fig. 1a), is surprisingly able to sustain droplet-jumping at elevated subcooling. The DOP-20 surface was fabricated on thin copper tubes (outer diameter 6.35 mm and inner diameter 4.00 mm) using an electrochemical deposition method with a deposition time of only 10 s, which is rapid, scalable, and highly-adaptive for curved surfaces, to generate the hierarchical porous structures, followed by hydrophobicization. Details concerning surface preparation and characterization can be referred to the supplementary material.

The surface superhydrophobicity is clearly exhibited by both the water repelling behavior and the static contact angle (see the insets in Fig. 1a). Using the focused ion beam technique, a DOP-20 sample was cut to show the lateral structures of the porous layer, whose thickness was measured to be ∼40 μm (i.e., the order of the valley floor). As shown in Fig. 1b, a smooth hydrophobic surface was prepared by a low surface energy coating, which was tested as the baseline case. In order to compare the spatial control effect through pore size variation, the deposition time was increased to 40 s to fabricate a contrast surface with pores having a larger nominal diameter of ∼50 μm and a greater depth of ∼80 μm (see Fig. 1c). However, as compared to the DOP-20 surface, the nanoscale morphology and static contact angle are almost unchanged on the DOP-50 surface.

Visualized condensation experiments were carried out to test the heat transfer performance of these surfaces over a wide range of subcooling up to 20 K. As shown in Fig. 2a, the copper tube was horizontally positioned in a vacuum chamber with observation windows. The chamber was evacuated to a pressure below 20 Pa to eliminate the non-condensable gases prior to each test, followed by injection of pure water vapor generated from an electrical boiler. The pressure in the chamber was maintained at ~8 kPa for all condensation experiments. The copper tube was connected to a water bath outside the chamber to build a cooling water loop at a set temperature and volume flow rate (280 L/h). The water inlet and outlet temperatures of the copper tube were monitored by two RTDs with high accuracy (±0.1 K). Except for the test section of a length of 150 mm, the other part of the copper tube and the auxiliary connections were insulated. With the help of an LED light, dynamic behaviors of condensate droplets during the condensation process was recorded using a digital camera equipped with a telephoto lens, which was synchronized with the heat transfer measurement.

As illustrated in Fig. 2b, the condensation heat transfer over the exposed copper tube was calculated based on the thermal resistance network analysis, where the overall thermal resistance was divided into three parts including the condensation of water vapor, heat conduction through the tube wall, and convection by the internal cooling water. The expressions for these thermal resistance terms are given in Eq. 2, where \( h_c \) stand for the condensation heat transfer coefficient, \( A_o \) the outer surface area of the tube, \( k_o \) the thermal conductivity of the tube wall (copper), \( h_i \) the internal heat transfer coefficient, \( A_i \) the inner surface area of the tube, and \( K \) the overall heat transfer coefficient. Because the latent heat released by vapor condensing on the tube led to temperature increase of the cooling water, the overall thermal resistance \( R \) was estimated by the enthalpy change in the cooling water. Then the condensation heat transfer coefficient \( h_c \) was able to be extracted by

\[
h_c = \frac{1}{A_o (R - R_w + R_i)}.
\]

Details concerning the experimental setup and procedure, data reduction, and uncertainty analysis can be referred to the supplementary material.

The typical condensate behaviors on these surfaces are compared at a relatively low surface subcooling of 4 K, as illustrated in Fig. 3. As expected, conventional dropwise condensation was achieved on the baseline hydrophobic surface on which the droplets shed off the surface under the force of gravity after growing to millimeter-sized, whereas coalescence-induced droplet jumping was realized with the use of the modified superhydrophobic surfaces (both DOP-20 and DOP-50). An intensive droplet jumping phenomenon can be clearly seen around the DOP-20 surface, as indicated by the densely-packed atomized droplets spraying from the tube (see Fig. 3b), despite the undesirable presence of sparsely-distributed pinned droplets that are much bigger (with diameter of the order of 100 μm–1 mm). From the zoom-in inset, the size of the tiny sparklets (i.e., the jumping droplets after coalescence) is estimated to be of the order of 10 μm, which is consistent with the size range of condensate droplets that are mostly possible to jump after coalescence (20–50 μm). Thanks to the keen-edged and serrated nano-structures on the hierarchical porous superhydrophobic surface.
surface (see the insets in Fig. 1a), the condensate droplets likely existed in the Cassie state, rather than the Wenzel state, that facilitates the occurrence of droplet jumping.\(^1\)

With increasing the nominal pore size of the hierarchical porous surface, droplet jumping can still be observed over the DOP-50 surface despite the deteriorated intensity of jumping, as evidenced by the presence of fewer sparklets in Fig. 3c. It was pointed out that in order to realize coalescence-induced droplet jumping there must be at least one relatively big droplet with the diameter exceeding 10 \(\mu\)m.\(^2\) Hence, the smaller pore size of \(\sim 20\) \(\mu\)m seems to be more appropriate for realizing more efficient coalescence among condensate droplets, due to the closer distance between neighboring droplets that nucleated and grew on the microscale ridges among the pores. This is also the reason why the pinned big droplets were also fewer on the DOP-50 surface, because there were more numbers of sub-10-\(\mu\)m pores on the DOP-20 surface (see Fig. 1a). These pores can easily trap droplets when they grow to the size of 10 \(\mu\)m without coalescence, finally leading to pinned millimeter-sized droplets.

The hypothesized scenario for droplet jumping on such porous surfaces at relatively low subcooling is illustrated by the schematics in the left column of Fig. 4, where the changes in the condensate behaviors at elevated subcooling is also presented. As shown in Fig. 4a, it can be seen that self-propelled droplet jumping was always able to be sustained on the DOP-20 surface with increasing the subcooling, in spite of the gradual decrease in the intensity of jumping. On the one hand, according to Eq. (1), with increasing the subcooling the critical nucleation size of the condensate droplets becomes smaller. On the other hand, the local pinning force becomes stronger at the liquid–solid contact regions at higher degrees of subcooling,\(^3\) leading to lower possibility to form Cassie-state droplets with high mobility.

As illustrated by the schematic on the right side in Fig. 4a, at a high surface subcooling, the condensate droplets nucleated in a smaller diameter and pinned more tightly with the structured surface which resulted in the difficulty in the droplet removal and the accumulation of bigger droplets on the surface, comparing to condensation at a low surface subcooling. In addition, non-condensable gases were likely trapped in the pores to serve as a thermal impedance to local condensation, indicating that the nanostructured ridge tips were able to facilitate the growth and removal of condensate droplets. From the droplet dynamics point of view, the microscale ridges served as spatial separation barriers that prevent the surfaces from nucleation-density-modulated flooding at high degrees of subcooling. Similar trends of the condensate behaviors can be seen for the DOP-50 surface with larger pore size and greater pore depth, as shown in Fig. 4b. With increasing the subcooling, the surface seems to be gradually wrapped with a white sheen layer, indicating that more condensate droplets were accumulated on the surface which were more difficult to be removed due to the larger pinning force between the droplets and surface, as compared to the case with a lower surface subcooling. However, droplet jumping can still be observed at the highest subcooling of 19 K despite a very weak intensity of jumping.
Having presented the condensate behaviors on these surfaces, the attention is now paid to their heat transfer performance. As compared to the baseline case of hydrophobic surface on which conventional dropwise condensation was able to attain, both the surface-averaged heat flux and heat transfer coefficient exhibit remarkable improvements on the DOP-20 surface for degrees of subcooling up to ~7 K (see Fig. 5). This suggests that enhanced condensation heat transfer was achieved successfully due to the realization of droplet jumping, which accelerated the removal frequency of condensate droplets. The dendritic nano-structures grown along random orientations on the modified surface (see Fig. 1a) may result in droplets jumping off the surface in different directions that increase the volume of condensates removed in each jump by sweeping the neighboring droplets. In addition, these dendritic nano-structures provided more sites for vapor clusters to nucleate, because of the lower interfacial free energy barrier comparing to that on the smooth hydrophobic surface. However, when the surface subcooling exceeds 7 K, the heat transfer enhancement due to fast removal of the coalescence-induced droplets jumping was unable to compensate the heat transfer impedance associated with the much thicker condensate film over the tube (due to the porous layer).

At the relative low subcooling of 4 K, there is a pronounced 3-fold increase of the surface-averaged heat transfer coefficient comparing to that of the smooth hydrophobic surface (see Fig. 5b), which is even slightly higher than that as measured by Miljkovic et al. on a superhydrophobic copper tube, having the same outer diameter and coated with blade-shaped CuO nano-structures, at the same surface subcooling. It is noted that the porous layer on the DOP-20 surface was thicker than the CuO coating in Miljkovic et al., indicating an additional thermal resistance was introduced. However, it can be easily estimated that the thermal resistance across the porous layer only accounts for a very small portion in the overall thermal resistance network (see Fig. 2b), so this additional thermal resistance seems to not deteriorate the heat transfer to a significant extent. The superior condensation heat transfer performance on the DOP-20 surface was likely attributed to the spatial separation effect associated with the porous structures that are hypothesized to reduce effectively the departing diameter of the jumping droplets, outweighing the additional thermal resistance of the relatively thick porous layer. It is also shown that the relative enhancement in heat transfer degraded significantly with increasing the subcooling, in accordance with the reduced intensity of droplet jumping. As expected, the heat transfer performance was deteriorated on the DOP-50 surface, and became worse than that on the smooth hydrophobic surface starting from a lower surface subcooling (~4 K). The even thicker porous layer on the DOP-50 surface as compared to the DOP-20 surface, meaning a greater additional thermal resistance when condensates were trapped in the pores, is likely attributed to this observation.

In summary, we have demonstrated that self-propelled droplet jumping mode of condensation was achieved on a superhydrophobic surface with hierarchical porous structures. Efficient droplet jumping was found to always sustain on the porous surface over a wide range of subcooling up to ~20 K, although enhanced heat transfer was only able to be achieved for up to ~7 K as compared to the conventional dropwise condensation on a smooth hydrophobic surface. Although porous surfaces with wicking ability has often been used to enhance filmwise condensation by lowering the effective thermal resistance across the condensate film, the specific superhydrophobic porous structures fabricated in this work, having ~20 µm pores with nanoscale dendritic morphologies, was able to facilitate coalescence-induced droplets jumping. Although a portion of droplets got trapped unfavorably in the pores, droplet jumping was able to be sustained at elevated subcooling due to the spatial separation effect of the microscale ridges among the pores.

The observations in this work shed light on a new way to achieve droplet jumping condensation at elevated subcooling by the use of porous surfaces. It was found, however, that the performance of such porous surfaces is very sensitive to the nominal pore size, because a match with the size of condensate droplets that are easy to coalesce for jumping should be reached. The electrochemical deposition method that was employed to fabricate the hierarchical porous surface is rapid (taking less than 1 min) and highly-adaptive for curved metallic surfaces, e.g., copper tubes, which exhibits a great potential for large-scale industrial applications involving condensation processes.

Details concerning the fabrication and characterization of the superhydrophobic porous surface, the test rig and procedure for the condensation experiments, as well as the data reduction and uncertainty analysis are made available in the supplementary material.

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