Boiling Heat Transfer with a Well-Ordered Microporous Architecture

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ABSTRACT: Boiling heat transfer through a porous medium offers an attractive combination of enormous liquid—vapor interfacial area and high bubble nucleation site density. In this work, we characterize the boiling performances of porous media by employing the well-ordered and highly interconnected architecture of inverse opals (IOs). The boiling characterization identifies hydrodynamic mechanisms through which structural characteristics affect the boiling performance of metallic microporous architecture by validating empirical measurements. The boiling performances can be optimized through the rational design of both the structural thicknesses and pore diameters of IOs, which demonstrate up to 336% enhancement in boiling heat-transfer coefficient (HTC) over smooth surfaces. The optimal HTC and critical heat flux occur at approximately 3–4 μm in porous structure thickness, which is manifested through the balance of liquid–vapor occupation within the spatial confinement of the IO structure. The optimization of boiling performances with varying pore diameters (0.3–1.0 μm) can be attributed to the hydraulic competitions between permeability and viscous resistance to liquid–vapor transport. This study unveils thermophysical understandings to enhance multiphase heat transfer in microporous media for ultrahigh heat flux thermal management.

KEYWORDS: boiling heat transfer, inverse opals, 3D microporous architecture, parahydrophobicity, wettability

1. INTRODUCTION

Boiling enhancements through modified surfaces promise performance breakthroughs for applications ranging from power generation to desalination and thermal management for high-power electronics and nuclear reactors.1–6 The latent heat of nucleate pool boiling promises up to 2 orders of magnitude higher in heat flux dissipation in comparison to single-phase liquid cooling.7 Extensive studies have explored various strategies to enhance boiling performances by investigating different types of surfaces or architectures. Planar surface modifications of surface wettability,8–12 roughness,12 and microstructures13–15 have demonstrated significant boiling heat-transfer improvements by increasing the liquid–solid interfacial area where thermal energy is converted to fluid enthalpy and by controlling bubble dynamics.16

A promising alternative to planar surface modifications for pool boiling enhancement is complex three-dimensional (3D) microporous architectures such as metallic foams, porous media, and sintered particles.17–19 These structures with highly interconnected pores provide an enormous solid–liquid interfacial area for enhanced thermal transport, capillary pressure for passive liquid wicking, and pathways for vapor to escape. The higher pore densities in those porous structures also increase the number of potential bubble nucleation sites. Such inherent structural characteristics of 3D microporous architectures significantly affect the pool boiling dynamics and can be tuned to optimize two-phase heat-transfer processes.

Despite intensive efforts within the past few decades, the fundamental understanding of 3D microporous coatings on boiling behaviors is still limited.20 The discrepancies are mainly due to the porous coating’s random pore arrangements and nonuniform pore size distributions,21 making it difficult to elucidate structure–property relations in pool boiling. Furthermore, the mechanisms by which the pore size affects the thermal and hydraulic instabilities associated with boiling remain unclear because of the lack of systematic investigations with uniform pore distribution. In the case of sintered particle coatings, the majority of studies report the particle size without specifying the dimensions of the interstitial cavities.22–25 This is most salient because particle sizes are not easily translatable to cavity dimensions. In the case of highly porous foam-like materials,18 the pore distributions and localized porosities widely vary.

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An emerging category of 3D microporous structure that is attractive for fundamental boiling investigation is inverse opals (IOs) because of the extreme structural regularity. IOs are created by inverting a template of self-assembled microspheres with a structural material to reveal a highly interconnected and porous scaffold. The IOs’ regular pore packing arrangement that derives from the crystalline self-assembly of monodispersed spheres can be exploited to systematically investigate pore characteristics on boiling behaviors. The additional capability to control the structural porosity and thickness also enables single-parameter studies by eliminating multiple interplaying thermo-fluidic mechanisms.

In this study, we investigate the pool boiling performance of 3D highly ordered nickel IOs in nucleate pool boiling conditions with deionized (DI) water at atmospheric pressure. To understand the thermophysical relationships between structural characteristics and boiling physics, we selectively modulate the porous media thickness (∼1 to 10 μm) and pore diameters (0.3 to 1.0 μm). The nickel IOs improve boiling critical heat flux (CHF) by up to 240% over flat surfaces where the peak boiling performance occurs at approximately 3 to 4 μm in structural thickness, prompted by the balance between the competing liquid–vapor interactions within the confinement of the pore network. We perform theoretical interpretations of the boiling phenomena inside highly ordered 3D interconnected microporous coatings by considering the interplay between dynamic wetting, evaporation, and viscous pressure drop associated with liquid–vapor ebullition cycle within the spatial confinement of the microporous matrix. The results are then compared to experimental CHF. This study aims to advance the fundamental insights toward boiling behaviors that can provide engineering guidance toward designing efficient two-phase thermal management devices for next-generation microelectronics.

2. RESULTS AND DISCUSSION

The efficacy of boiling can be quantified by the heat-transfer coefficient (HTC), which is defined as the applied heat flux divided by a given temperature difference between the heated surface and bulk fluid (denoted as the wall superheat ΔT = Twall − Tsat). The HTC improves with increasing heat flux until the CHF is reached, where the departing bubbles coalesce to form a thermal vapor barrier between the solid surface and bulk liquid. Such conditions significantly reduce the heat transfer from the hot solid to the liquid coolant as observed by the drastic increase in surface temperature. The surface burnout at CHF therefore indicates the boiling heat-transfer performance limit.

We assess the boiling performances of our nickel IO structures using the resulting HTC values and compare them to other types of 3D interconnected porous structures for a wide range of pore sizes ranging from ∼0.3 to 800 μm. The microporous architectures utilized by other investigators include sintered particles, electrodeposited metal around electrochemically generated bubbles, nanoporous alumina, and commercially available metallic foams. While these classes of porous structures all possess random pore interconnectivity and relatively large pore size disparities, they are evaluated owing to their similar structural characteristics to that of IOs: 3D pore interconnectivity with definable pore sizes. In contrast, micropillars and nanowire arrays are not considered for comparisons in this study because they lack such structural characteristics. The pool boiling measurements from these previous studies are also comparable to our work because of similar experimental conditions: DI water as the working fluid and experimentation under atmospheric pressure. Because the boiling measurement setup varies between investigative groups, the boiling performances may slightly differ (see the Supporting Information for more details), normalizing that the reported maximum HTC values of the porous coatings to those of the plain surfaces enables appropriate comparison in the heat-transfer enhancements contributed by the microporous surfaces, as shown in Figure 1.

![Figure 1. Comparing the boiling performances of prior 3D interconnected porous structures. The highest reported HTCs from previous investigations are normalized to that of their respective plain surfaces, which are then compared to the boiling enhancement reported from this study (highlighted in yellow).](https://dx.doi.org/10.1021/acsami.0c01113)

Prior studies demonstrate between approximately 150 and 315% in maximum HTC enhancement. Details regarding the CHFs and HTCs reported in these studies are presented in Table 1, and it should be noted that only the highest HTC of monolithic porous coatings is considered from each study. In comparison, our nickel IOs improve boiling by 336 and 303% in HTC for 0.6 and 1.0 μm pore diameter structures, respectively, over plain silicon. While the maximum HTC enhancement for porous coatings is presented in Figure 1, the boiling performances for our nickel IOs can vary widely (between ∼100 and 300% HTC improvement) depending on their structural thicknesses and pore diameters. These variations in boiling performances beckon further examination to reveal structure–performance relations (i.e., structural thickness and pore diameter dependence) in well-ordered interconnected microporous architectures.

In this study, we first characterize the surface wettability of nickel IOs periodically under their boiling condition to understand the transient changes in surface energy and their resulting effects on the liquid–vapor transport behaviors. We then present the traditional understandings of liquid–vapor ebullition behaviors within porous media, followed by thermophysical predictions of boiling performances based on fundamental hydrodynamic principles. The thickness-dependent boiling performances are evaluated by experimentally measuring the CHF and HTC of variable IO thicknesses for two sets of pore diameters (0.6 and 1.0 μm). We further examine the pore diameter-dependent boiling performances to gain insights into the effects of permeability on vapor extraction during boiling.

2.1. Surface Wettability. For hydrophilic porous media, the hydrodynamic principles during an ebullition cycle presented in previous studies explain a continuous liquid wicking driven by the pore capillary forces to rewet the boiling
surfaces. This argument assumes that the surface energy of the microporous medium remains constant throughout the boiling process (i.e., highly wetting), which has been rarely validated, especially for highly conductive metallic surfaces (e.g., copper and nickel) that are prone to oxidation and changes in surface energy. To provide insights toward the transient surface wettability during pool boiling, we measure the water contact angle of the nickel IO surface before and periodically over a span of 3 h under boiling conditions. The surface wettability of the nickel IOs prior to the boiling measurements shows hydrophilic characteristics with receding and advancing contact angles of $\theta_{\text{rec}} \approx 15^\circ$ and $\theta_{\text{adv}} \approx 40^\circ$, respectively (Figure S1). The relatively hydrophilic surface rapidly increases in hydrophobicity with $\theta_{\text{adv}} \approx 110^\circ$ after the first hour of boiling, before stabilizing at $\theta_{\text{adv}} \approx 120^\circ$ after 3 h (Figure S2). The changes in surface wettability can be attributed to surface chemistry modifications caused by metallic oxidation under boiling conditions because no physical changes in the porous features are observed before and after the boiling process (Figure S2).\textsuperscript{34,35} Because the advancing contact angle quickly increases to $\sim 120^\circ$ within a few hours, the nickel IOs are assumed to be nonwetting during the entirety of the boiling experiments. However, it is noteworthy to mention that the receding contact angles $\theta_{\text{rec}}$ remain consistently low at $\sim 20^\circ$ before and during the boiling experiment. This suggests strong liquid impingement upon the microporous surfaces, which is consistent with our previous finding that demonstrated water droplets remaining pinned to nickel IOs even when the surface was tilted to $90^\circ$, despite having a hydrophobic static contact angle of $130^\circ$ irrespective of pore diameter or thickness.\textsuperscript{36} The wetting characteristics of nickel IOs presented here resemble those of parahydrophobic surfaces (i.e., high static contact angle and high contact hysteresis).\textsuperscript{37} The strong adhesion of the droplet to the IOs indicates a Wenzel wetting state, such that the droplet sinks into the microporous surface.\textsuperscript{38} In this dynamic wetting state, the three-phase contact line remains pinned to the surface and bubbles grow with a fixed base diameter, mitigating vapor spreading and delaying CHF. See Figure S3 for representative bubble growth and departure characteristics during the ebullition cycle. Before the incipience of boiling, the liquid from the pool reservoir impinges and fills the cavities of the IOs across their thickness, displacing the entrapped air through their highly interconnected cavities and effectively wetting the surfaces. During nucleate boiling, the Wenzel wetting state can promote the replenishing liquid to penetrate through the pore spaces left behind by the escaping bubbles by the Laplace pressure. The liquid rewetting mechanism promoted by the Wenzel state presented here is evident from the observed boiling behaviors. That is, even though hydrophobic and superhydrophobic surfaces can initiate boiling incipience more efficiently at lower heat fluxes, their inability to rewet the surfaces quickly leads to premature film boiling at moderate to high heat fluxes. However, the sustained nucleate boiling of our hydrophobic nickel IOs strongly suggests that the surface energy is capable of maintaining a stable liquid film inside the porous structure.

2.2. Bubble Behaviors in Porous Medium. In pool boiling, bubble dynamics are characterized by the cyclic mechanisms of liquid replenishment and vapor generation. That is, liquid rewets the phase-change surfaces as vapor bubbles grow and depart from the heated surface. The liquid contributes to phase change through evaporation and subsequent bubble nucleation, as illustrated in Figure 2. The ebullition cycle continues at relatively low heat fluxes until the rate of bubble nucleation and departure begins to outpace that of liquid rewetting at the CHF.

The bubble growth and departure dynamics are observed using high-speed recording during boiling incipience. As
bubbles nucleate and grow, their contact line spreads outward and the liquid recedes away, which closely corresponds to the contact angle of a receding droplet at the early stages of bubble growth (when buoyancy exhibits minimal distortion effect on the bubble shape). Therefore, the bubble growth characteristics can provide insights into the wettability of the boiling surfaces. For our nickel IO surfaces, both the bubble base diameter and the liquid receding contact angle remain small during bubble growth. This inhibits the spread of vapor across the surface and allows the surfaces to continue undergoing nucleate boiling. The growing bubble then departs from the surface once it reaches the critical size where buoyancy forces exceed the surface tension forces. See Figure S4 for details on the bubble departure diameters and departure frequencies near boiling incipience.

Below the base of the growing bubble, evaporation of thin liquid film contributes to the bubble growth and phase-change heat transfer. When the liquid completely saturates all the available pores and evaporation occurs at the top of the porous structure, the evaporation rate reaches its maximum. Within the thickness of the IO structure, the general region filled with liquid where evaporation is most likely to occur can thus be denoted as the “evaporation regime”, as illustrated in Figure 2. However, the liquid permeation through the porous media (even under the Wenzel dynamic wetting state) during boiling is often nonuniform and unpredictable because of the violent bubble extraction through the tortuous confinement of 3D microporous media. That is, the occupation of vapor across multiple interconnected pores can also block permeable pathways for liquid replenishment. The effect of nonuniform liquid rewetting on boiling performance is especially poignant with increasing microporous media thickness. While increasing the microporous media thickness increases the interfacial surface area for a higher phase-change rate, beyond a certain optimal thickness can cause the escaping vapor to travel a greater distance across the thickness of the microporous media, during which the bubbles continue to grow and occupy more of the interconnected pore cavities, further limiting the liquid-permeable pathways and increasing the thermal resistance across the IO thickness. The partial dry out caused by vapor pockets consisting of large bubbles will mostly occur near the base of the microporous media where the heat source is the greatest and can generally be deemed as the “boiling regime”, as illustrated in Figure 2. The complexity of boiling within 3D microporous architectures can be further elucidated by modeling the liquid—vapor interfacial transport behaviors as a function of structural thickness.

2.3. Theoretical Evaporation and Vapor Transport Limits. Advection heat transfer dominates as liquid impinges and permeates through the heated surfaces of the porous medium near the top porous surface adjacent to the pool reservoir. In addition to surface advection, the evaporation between liquid—vapor interfaces contribute to the overall heat and mass transfer because of liquid—vapor phase change in pool boiling. The evaporation of the liquid can estimate the overall CHF, \( q_{\text{CHF, evap}}^* \), by accounting for the baseline boiling heat-transfer contribution from the plain surface represented as \( q_{\text{CHF, plain}}^* \) as well as the enhancement gain from the microporous coating denoted as \( q_{\text{evap}}^* \)

\[
q_{\text{CHF, evap}}^* = q_{\text{CHF, plain}}^* + q_{\text{evap}}^*
\]

When the impinging liquid evaporates and contributes to the formation of vapor bubbles, the maximum evaporation rate per bubble in the evaporation regime can be described as

\[
M_{\text{evap}} = \pi d_b \int_0^\delta M_{\text{evap}}^* \rho d \delta
\]

where \( \varphi \) is the structural porosity, \( \delta \) is the thickness of the porous structure, \( d_b \) is the diameter of the bubble contact line with the porous surface, and \( M_{\text{evap}}^* \) is the evaporation mass flux between the liquid—vapor interfaces within the porous medium. The derivation of \( M_{\text{evap}}^* \) assumes that evaporation occurs at a stationary liquid—vapor interface maintained by steady-state heat conduction through the porous medium. The resulting CHF contributed by evaporation occurring near the bubble formation at the top of the IOs can be estimated as

\[
q_{\text{evap}}^* = \frac{M_{\text{evap}}^* h_{fg}}{2d_b^2}
\]

where \( h_{fg} \) is the latent heat of vaporization.

The evaporation of liquid to form vapor bubbles can also occur within the porous medium as the wall temperature exceeds the saturation temperature. As the nucleated vapor escapes upward from the base of the media, the bubble exudes a maximum viscous pressure loss \( \Delta p_v \) on the system that can be estimated using Darcy’s law as

\[
\Delta p_v = \frac{\mu \delta U_v}{K_v}
\]

where \( \mu \) is the vapor dynamic viscosity, \( \delta \) is the structural thickness, and \( K_v \) is the vapor permeability through the porous medium. Our computational fluid dynamics (CFD) simulation shows that the vapor permeability \( K_v \) through a unit cell of IO is an order of magnitude higher than liquid permeability \( K_l \) for a given pore size distribution (see Figure S5 for calculation details). Even so, vapor transport is often hindered by partial liquid—vapor blockages within the microporous structure. As a result, the mixture of liquid—vapor transport becomes limited by the lower liquid permeability \( K_l \) before approaching complete dry out, and by equating \( K_v \) to \( K_l \), we can more accurately represent the boiling limit. The vapor blockage often occurs near the base of the microporous structure where the heat flux is the highest. As the vapor moves upward through the porous medium with uniform pore diameter \( d_p \), the vapor velocity, \( U_v \), can be determined as

\[
U_v = \frac{q^*}{h_{fg} \rho_v}
\]

where \( \rho_v \) is the vapor density.

Below the CHF in pool boiling, the transport of the bubble through the porous medium causes a viscous pressure loss, which is assumed equal to the increased vapor pressure experienced at the wall superheat. The vapor pressure drop \( \Delta p_v \) can be estimated over the wall superheat \( \Delta T \) to express the pressure—temperature relation along phase boundaries, using the Clausius–Clapeyron equation

\[
\frac{\Delta p_v}{\Delta T} = \frac{h_{fg}}{T_{sat} \Delta v}
\]

where \( \Delta v \) is the difference in specific volume between liquid and vapor phases, represented as
where $\rho_l$ is the liquid density, and $T_{sat}$ is the saturation temperature, set equal to 372 K. The combination of eqs 5 and 6 estimates the CHF by considering the vapor transport through the porous medium with a determined structural thickness $\delta$. The boiling performances initially increase for both sets of pore diameters until reaching a critical thickness, which then causes the boiling performances to decrease. The evaluated samples possess a porosity of 80%.

$$\Delta v = \frac{\rho_l - \rho_v}{\rho_l \rho_v}$$  \hspace{1cm} (7)$$

where $\rho_l$ is the liquid density, and $T_{sat}$ is the saturation temperature, set equal to 372 K. The combination of eqs 5 and 6 estimates the CHF by considering the vapor transport through the porous medium with a determined structural thickness $\delta$. The structural porosity $\phi$ of the IO film is maintained at 78–80% (see Experimental Section for porosity calculations). To demonstrate the heat-transfer enhancement contributed by the IO surfaces, their boiling performances are normalized to those of polished silicon surface (CHFSi = 47 W/cm$^2$ and HTCSi = 2.11 W/cm$^2$K). The boiling curves for polished silicon are shown in Figure S6. The HTC is the maximum value across the heat flux ranges, which is often observed near the CHF. We note that the CHF of our polished silicon is lower than those presented in the literature, which is typically CHFSi = 66–89 W/cm$^2$ and HTCSi = 1.66–3.89 W/cm$^2$K for the same boiling conditions: water as the working fluid at atmosphere pressure (see Table S1 for details).42–46 We believe that our discrepancy from literature values is based on how the sample is mounted onto the heater, which varies between studies, and we have demonstrated that these variances in mounting configurations can alter the heat-transfer performance (see Figure S6 and Table S2 for details). Nevertheless, because our HTCSi is within the range of prior

Figure 3. Structural thickness-dependent pool boiling performance. Cross-sectional SEM images of nickel IOs with varying structural thicknesses for pore diameters of (a) 0.6 and (b) 1.0 $\mu$m. The boiling curves of (c) 0.6 and (d) 1.0 $\mu$m pore diameter nickel IOs with thicknesses ranging from ~1 to 10 $\mu$m. The horizontal arrows indicate the initiation of CHF. HTC as a function of heat flux for (e) 0.6 and (f) 1.0 $\mu$m pore diameter nickel IOs. The boiling performances initially increase for both sets of pore diameters until reaching a critical thickness, which then causes the boiling performances to decrease. The evaluated samples possess a porosity of 80%.
works, we expect that by normalizing the HTC of nickel IOs to that of bare silicon, we can provide a basis for reasonable comparisons between the boiling efficacy of different 3D porous media (as previously shown in Figure 1). The boiling curves for structural thickness-dependent IOs are presented in Figure 3c–f, which shows that both the CHF and HTC initially increase with increasing structural thickness until the optimal thickness ($\delta = 3$–$4$ $\mu$m). Further increases in sample thickness lead to a decrease in both CHF and HTC. Such general trends for both sets of pore diameters suggest that boiling performance can be optimized by selecting the appropriate structural thickness for each pore diameter. In terms of HTC, the optimal thicknesses for 0.6 and 1.0 $\mu$m pore diameters are 4 and 3 $\mu$m, respectively, yielding 7.13 and 6.39 W/cm$^2$K. In terms of CHF, the optimal thickness is ~3 $\mu$m for both pore diameters (113.0 and 97.5 W/cm$^2$ for 0.6 and 1.0 $\mu$m pore diameters, respectively). The optimum thickness represents the balance between competing hydrodynamic liquid–vapor behaviors that support efficient removal of heat fluxes from the microporous surfaces.

The evaporation limit (eq 1) and vapor transport limit (eq 8) are plotted as a function of structural thickness $\delta$ and overlaid on top of the experimental CHF of the 0.6 and 1.0 $\mu$m pore diameter nickel IOs (Figure 4). The calculation of the evaporation limit assumes that the liquid infiltrates the entirety of the IO thickness and suggests that CHF enhances with increasing $\delta$. However, for the experimental data, the CHF maxima and reversal in boiling performance with higher $\delta$ are prompted by the vapor transport limit, which considers the liquid–vapor interfacial transport inside the IOs. The wall superheat value $\Delta T$ used in eq 8 is the average $\Delta T$ from various $\delta$ values studied in their respective set of $d_p = 0.6$ and 1.0 $\mu$m. The theoretical limits predicted using fundamental hydrodynamic principles match relatively well with the experimental data. This can be contributed to the highly ordered microporous architecture of IOs with explicitly controllable features.

The departing bubble diameters and departing frequencies are measured from high-speed recordings near the incipience of boiling. A representative camera captures the bubble characteristics and results are shown in Figure S4. The reported bubble diameter and departing frequency are measured from nucleation sites near the center of the boiling surface and are observed from bubbles that grow and depart without coalescing with the surrounding bubbles during the ebullition cycle. The improved boiling HTCs shown approaching the optimal structural thickness are supported by the release of smaller-bubble diameter at faster departing frequency. The reversal in boiling performance with increasing thickness is attributed to the release of increasing bubble diameter at a slower rate.

It should be noted from Figure 4 that the 1.0 $\mu$m pore diameter IOs consistently exhibit lower boiling performances than the 0.6 $\mu$m pore diameter IOs for any given $\delta$. To further understand this phenomenon, we extend our investigation to study the effect of IO pore diameter on boiling performance.

2.5. Pore Diameter dependency. The boiling performances of nickel IOs with varying pore diameters ($d_p = 0.3, 0.6,$ and 1.0 $\mu$m) are examined, as presented with their corresponding SEM images in Figure 5. The IO films maintain a constant $\delta = 2$ $\mu$m and $\varphi = 80\%$ to isolate the pore diameters’ effects on boiling performances. The resulting boiling curves are presented in Figure 5b, showing the CHF increases from 48.8 to 61.2 W/cm$^2$ for $d_p = 0.3$ and 0.6 $\mu$m, respectively. Further increasing $d_p$ to 1.0 $\mu$m causes the CHF to decrease to 35.4 W/cm$^2$. This may be unexpected because larger $d_p$ provides higher structural permeability for improved liquid–vapor transport. For instance, we can analytically estimate the liquid permeability $K_l$ by relating a constant structural porosity $\varphi$ and variable pore diameters $d_p$ through an expression $K_l = d_p^{-2}(0.07\varphi^2 - 0.0539\varphi)$, derived using CFD simulation results and is valid for $78\% < \varphi < 92\%$. With increasing $d_p$ from 0.3 to 1.0 $\mu$m, $K_l$ of IOs is expected to increase from $1.5 \times 10^{-16}$ to $1.7 \times 10^{-12}$ m$^2$. However, the enlargement of $d_p$ may also contribute to a decrease in viscous resistance that can begin to dominate hydraulic transport physics. The lower viscous resistance exacerbates the competition between vapor and liquid transport. That is, in the liquid–vapor counterflow, the growing vapor pockets can be suppressed inside the IOs longer by the replenishing liquid before the vapor can escape. This in turn causes the boiling regime to grow and choking the liquid–vapor transport. This is evident when comparing the bubble extraction rate of $d_p = 0.6$ $\mu$m to 1.0 $\mu$m samples with varying $\delta$ from Section 2.4. In comparison to the bubble departure frequency of 1.0 $\mu$m pore diameter IOs, the 0.6 $\mu$m pore diameter IOs nucleate and depart smaller bubbles at a faster frequency (Figure S4), which helps maintain a cooler surface temperature and promotes efficient vapor removal and therefore enhancing the rate of heat flux removal.

The HTCs with increasing $d_p$ shown in Figure 5c show a similar trend to that of the CHFs with a maximum at $d_p = 0.6$ $\mu$m. We plot the calculated liquid permeability (blue line) as a function of $d_p$ in Figure 5d, which demonstrates the assistance of liquid–vapor routing through the enlarged pathways. The viscous resistance to fluid flow scales inversely with the square of $d_p$ and is plotted as the purple dashed line. The negative relationship of viscous resistance with increasing $d_p$ may
explain the general decrease in the boiling performance for porous structures with larger \(d_p\), as shown in Figure 1. However, more effort is needed to confirm such relationships by examining IOs with a wider range of \(d_p\) (>1 \(\mu\)m). Similar to rationally designing future phase-change devices with optimal structural thicknesses, the ideal pore diameter must also be considered to enhance the heat-transfer performance.

2.6. Structural Durability of Porous Surfaces. Last, to provide application-driven perspectives on future phase-change technologies, it is imperative to examine the feasibility of utilizing metallic IO coatings for two-phase heat transfer. In order to consider the durability and performance repeatability of IO coatings, we investigate the structural changes endured by the nickel IOs as a result of boiling. Here, we show that the physical morphology of the nickel IOs remains unchanged before boiling and after reaching CHF (see SEM images in Figure S2). This contrasts with copper IOs previously used in boiling heat-transfer study, which reports the destruction of the IO architecture with increasing boiling exposure due to surface oxidation.\(^{35}\) The superior capability of nickel to maintain its structural integrity suggests that nickel serves as an ideal architectural material for IOs because of its relatively high thermal conductivity and limited oxidative states. From an objective standpoint, the analysis of nickel durability presented here reiterates the imperative of material selection suitable for the intensive hydraulic and thermal environments of pool boiling.

3. CONCLUSIONS

Utilizing the well-regulated structure of IOs, we elicit new insights toward the fundamental boiling behaviors in 3D highly interconnected metallic microporous media. The uniformity of the IO architectures allows us to systematically modulate the structural features and examine their mechanistic role in triggering the onset of CHF. By rationally designing the IO structures with specific combinations of structural characteristics (e.g., structural thickness and pore diameter), boiling HTC can be optimally enhanced up to 336% in comparison to smooth silicon surfaces. With variable structural thicknesses, we consider the hydrodynamic competition between the evaporation of the impinged liquid and the viscous resistance for vapor to escape within the spatial confinement of the microporous matrix. The increases in heat flux causes vapor pockets to form near the base of the heater and impede liquid flow to the phase-change surfaces, denoted as the boiling regime. While these vapor pockets contribute to improved bubble departure and consequently heat removal, the increase in structural thickness allows the vapor growth to spread within the porous media and cause severe localized dry outs. The balance between the hydrodynamic liquid–vapor behaviors within the confinement of a rationally designed microporous structure thickness can lead to optimal boiling heat transfer.

For a given structural thickness, the increasing pore diameter can enhance the boiling performances such as CHF and HTC because of the enlargement in permeability for improved liquid–vapor routings, but the continual increase in pore sizes can also limit boiling heat-transfer improvement because of the decrease in the viscous resistance to fluid flow, exacerbating the competitions between liquid–vapor counterflow.

In addition, the surface wettability of nickel IOs is examined over the duration of the boiling experiment to understand its effect on the intrinsic liquid rewetting of the surfaces. While
the nickel IOs are hydrophilic before boiling, they quickly transitioned to being hydrophobic within the first hour of the experiment by exhibiting high static and advancing contact angles. However, the low receding contact angles suggest that the liquid remains strongly pinned to the porous structure in a Wenzel wetting state. The strong propensity to impinge liquid may contribute to the rewetting of the boiling surfaces and prevent premature film boiling often observed for hydrophobic surfaces.

Studying the wetting durability of materials is critical to the understanding of the dynamic wetting behaviors in a complex hydrodynamic phase-change process, which warrants closer examination for future boiling heat-transfer investigations. The fundamental interfacial and structure—thermophysical relations elucidated from this study can significantly contribute to the design of next-generation phase-change thermal management devices.

4. EXPERIMENTAL SECTION

4.1. Nickel IO Preparation. Nickel IOs are fabricated by templating self-assembled spheres (see Figure S7 for process schematics).29 In this approach, a double-sided gold-coated silicon substrate is functionalized in 1 mM aqueous sodium 3-mercaptop-1-propanesulphonate for at least 24 h to render the surface hydrophilic. The substrate is vertically submerged in a colloidal suspension (0.6% water and further treated with plasma cleaning (Gatan, Solarus Model 92697, United States; orcid.org/0000-0002-8838-6213; Email: won@uci.edu)

4.3. Structural Porosity Calculation. Top and cross-sectional images of the IO are obtained using a scanning electron microscopy (FEI Quanta) and postprocessed with ImageJ (NIH). The diameter measurements of the via \( d_{\text{via}} \) and pore \( d_p \) are used to determine the IO structural porosity through a numerical correlation derived using CFD models described in previous work.29,30 \[ \phi = 0.5833(d_{\text{via}}/d_p) + 0.6633. \]

Rigorous measurements of \( d_{\text{via}} \) and \( d_p \) are collected, and representative statistical distributions for each set of pore diameters are shown in Figure S8.

4.4. Pool Boiling Measurement. Nucleate pool boiling heat transfer of nickel IO-coated surfaces is experimentally investigated with varying pore diameters and structural thicknesses. DI water serves as the working fluid, which is degassed through continuous boiling for \( \sim 30 \) min before experiments. At the base of the apparatus, four cylindrical cartridge heaters (Omega, CIR-2015) controlled by an ac voltage regulator (Variac Transformer) heat an encompassing copper block to transport heat flux toward the sample. A thermocouple located slightly above the heated sample is connected to a guard heater through a proportional integral derivative controller, which ensures that the pool liquid temperature remains saturated. By modulating the electric power supply through the cartridge heaters at the base, the applied heat flux of the copper block can be slowly increased. At each heat flux, temperatures are monitored through a linked data acquisition system (LabJack U6) and recorded in steady state to produce the boiling curves. More details regarding heat flux calculations are provided in the Supporting Information. The accuracy of the K-type thermocouple is within \( \pm 0.1 \) °C. By using the law of propagation of uncertainty, the maximum uncertainties of heat flux and HTC are \( \pm 7.26 \) W/m² and \( \pm 10.28 \) °C, respectively. A clear boiling chamber is used to allow visual monitoring of nucleating bubble dynamics (see Figures S9 and S10 for schematics for boiling apparatus and sample mounting as well as Table S3 for calculation details). A high-speed camera (FASTCAM Mini AX50) captures the dynamics of bubble nucleation and departure at 2000 fps through the transparent wall of the boiling chamber at 1024 \( \times \) 1024 pixel resolution. The departing bubble diameters and maximum base diameters are measured from unobscured nucleation sites that do not coalesce with other sites during the bubble growth and departure cycle.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsami.0c01113.

Pore diameter-dependent surface wettability, nickel IO structure characteristics from boiling, bubble growth and departure characteristics on hydrophobic nickel IOs, bubble characteristics and dynamics, water vapor permeability through IO calculations, liquid and water vapor permeability, boiling performances of plain silicon as reference surface, variances in sample mounting techniques, preparation of nickel IOs, representative statistical distribution of pore and via diameters, schematics of pool, boiling experimental setup, schematic of sample mounting, heat-transfer calculations, and film characteristics for pool boiling calculations (PDF)

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Notes
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