Ultrahigh Flux Thin Film Boiling Heat Transfer Through Nanoporous Membranes

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ABSTRACT

Phase change heat transfer is fundamentally important for thermal energy conversion and management, such as in electronics with power density over 1 kW/cm². The critical heat flux (CHF) of phase change heat transfer, either evaporation or boiling, is limited by vapor flux from the liquid-vapor interface, known as the upper limit of heat flux. This limit could in theory be greater than 1 kW/cm² on a planar surface, but its experimental realization has remained elusive. Here, we utilized nanoporous membranes to realize a new “thin film boiling” regime that resulted in an unprecedentedly high CHF of over 1.2 kW/cm² on a planar surface, which is within a factor of four of the theoretical limit. The liquid supply is achieved through a simple nanoporous membrane that supports the liquid film, with its thickness automatically decreases as heat flux increases. The thin film configuration reduces the conductive thermal resistance, leads to high frequency bubble departure, and provides separate liquid-vapor pathways, therefore significantly enhances the heat transfer. Our work provides a new nanostructuring approach to achieve ultrahigh heat flux in phase change heat transfer and will benefit both theoretical understanding and application in thermal management of high power devices of boiling heat transfer.
**KEYWORDS:** Thin film boiling, kinetic limit, ultrahigh flux, critical heat flux, nanoporous membrane

**TOC GRAPHIC**
Liquid-vapor phase change heat transfer is fundamentally important for a variety of processes, such as power generation, refrigeration, water desalination and purification, and thermal management of high-power electronics\textsuperscript{1-8}. Boiling is considered a promising cooling technique compared with other liquid-vapor phase change cooling phenomena such as jet impingement, spray cooling and thin film evaporation, which either have high operating temperature or have yet to be developed to achieve high heat fluxes\textsuperscript{9,10}. Both flow boiling and pool boiling has been studied extensively to enhance both critical heat flux (CHF) and heat transfer coefficient (HTC)\textsuperscript{4}.

In pool boiling, a heated surface is submerged inside a stagnant liquid pool, and the nucleation and subsequent bubble growth and departure carry the heat away from the heater. As a result, the CHF and heat transfer behaviors for pool boiling are largely dictated by the bubble dynamics. There are two different limiting cases associated with bubble evolution in pool boiling\textsuperscript{11}. When the bubbles are small, the bubble growth is in the inertia-controlled regime where the growth is rapid and is limited by the inertia force of the surrounding liquid, and all the interface area has large superheat. When the bubble size increases and exceeds the thermal boundary layer thickness as shown in Figure 1a, the further growth of the bubble is in the heat-transfer-controlled regime, where the growth is slow due to the depleted superheat at the liquid-vapor interface. While it is evident that the inertia-controlled regime is more desirable for more efficient heat transfer due to the faster bubble growth, it is difficult to prevent the transition into the heat-transfer-controlled regime as the bubbles grow. Therefore, pool boiling typically has low CHF, e.g., \~100 W/cm\textsuperscript{2} for water at the atmospheric pressure. Various strategies have been
explored to study the bubble dynamics and enhance pool boiling CHF\textsuperscript{12-18}, but the CHF still remains below 250 W/cm\textsuperscript{2}.

Flow boiling, on the other hand, can utilize fluid pumping to facilitate the bubble dynamics, as the liquid coolant is forced to flow along heated channels and the bubbles can be removed by the external pumping force. Therefore, flow boiling often has a higher CHF compared to pool boiling, especially in microchannels having small hydraulic diameter and consequently a large surface area-to-volume ratio. Recently, microchannel flow boiling devices with cooling flux close to or above 1 kW/m\textsuperscript{2} has been reported with different channel configurations\textsuperscript{19-21}. Other heat sinks\textsuperscript{22, 23} implementing boiling heat transfer using three-dimensional (3D) geometry also show CHF over 1 kW/cm\textsuperscript{2}. In these cases, the channels, grooves and pin fins made of high thermal conductivity materials (such as Si, Cu and diamond) are the key to spread the ultrahigh heat flux from the heater area to the extended surface area, thereby effectively reducing the heat flux on the actual phase change area to be well below 1 kW/cm\textsuperscript{2}.

Despite these innovative 3D device designs achieving high CHFs, an interesting question still remains: what would be the upper limit of the heat flux in boiling heat transfer from a planar surface? The answer to this question would not only have fundamental significance on exploring the limit of phase change heat transfer but could have implications on future development of high power thermal management technologies as any enhancement on planar surface can be implemented into 3D architectures. In this work, we utilized a simple nanoporous membrane structure that exploits the benefits of both flow boiling and pool boiling, and realized a new boiling scheme, termed “thin film boiling”, with an unprecedentedly high CHF on a planar
surface. As shown in Figure 1b, the boiling occurs on the heated surface of a nanoporous membrane, and the resulting bubbles carry the heat from the solid surface to the vapor space, as in the case of pool boiling. However, instead of having an extensive liquid pool, the liquid film is very thin and spontaneously becomes thinner as heat flux increases, which results in a reduced thermal resistance for conduction and renders the entire bubble growth process within the inertia-controlled regime. Consequently, the bubble departure size is small due to the constraint of the liquid film thickness, which promotes the bubble departure. Further, the liquid is supplied from the nanoporous substrate through liquid pumping akin to flow boiling, rather than from the lateral liquid replenishing mechanism as in typical pool boiling. Therefore, separate liquid-vapor pathways\textsuperscript{24,25} can be created for better heat transfer performance.

We experimentally investigated the boiling heat transfer performance of this proposed configuration using de-ionized water (DI water) as the working fluid and nanoporous anodic aluminum oxide (AAO) membranes as the porous substrates (Figure 2a,b). Water was driven across the membrane by pressure difference between the liquid and vapor sides of the membrane (Figure 2c). We then compared the measured CHF to the theoretical upper limit of phase change heat flux from a planar surface.

Commercial AAO membranes with nominal pore size of 200 nm and thickness of 60 µm were characterized with scanning electron microscope (SEM) as shown in Figure 2a, b, for the top view and cross-sectional view, respectively. The membrane was attached onto a PMMA sample holder with a $0.71 \times 0.71$ cm\(^2\) (0.50 cm\(^2\)) square hole with blade casted Torr Seal® Low Pressure epoxy. After epoxy curing, a $\sim 70$ nm thick square Pt layer with the same size as the
holder opening and two 2 µm thick Cu contact pads connected to the heater were deposited by sputtering (Figure 2c zoomed-in view). The exact sizes of the Pt layer and the Cu pads were precisely determined by shadow masks fabricated using a computer numerical controlled (CNC) milling machine. A 10 nm thick Cr layer was sputtered and served as an adhesion layer before both Pt and Cu deposition processes. The Pt layer served as both a heater and a resistance temperature detector (RTD).

After the deposition of the Pt heater and the Cu contact pads, the PMMA sample holder attached with the AAO membrane was assembled onto a custom-made liquid supply channel using screws and O-ring sealing. The low thermal conductivity of PMMA ensures good thermal insulation and minimizes conduction heat loss (Supporting Information S3). To make good electrical contacts to the Cu pads, thick Sn foils were pressed against the contact pads using custom-made PMMA clamps and set screws, as shown in Figure 2c.

During the experiments, the pressures of both the vapor side (top) and liquid side (bottom) of the membrane were controlled and maintained at preset values. The heating power was applied to the sample through direct current (DC) power supply to the Pt heater incrementally and the corresponding temperature was determined by the Pt RTD. The heat flux reported in this study was the actual heating power divided by the physical area of the AAO membrane (0.5cm²). Boiling experiments were performed under variable liquid pressure ranging from 252 to 1395 torr, while the vapor pressure was maintained at 17.5 torr (20 °C saturation temperature for water) for all the tests. Detailed experimental procedure can be found in Supporting Information.

Our experiment is a hybridization of flow boiling and pool boiling, as the liquid (water) was
pumped through the porous AAO membrane using the pressure difference between the liquid pressure ($P_1$) and vapor pressure ($P_r$) (Figure 2c), but the pumping was merely used to supply the liquid to the thin film liquid pool covering the heater deposited on the membrane (Figure 1b). As we shall see later, the membrane was covered with a liquid film throughout the entire experiment, so the phase change surface area was the same as the heater area. Figure 3 shows the boiling curves, or heat flux versus the superheat. Here the heat flux is defined as the applied heating power divided by the physical area of the membrane ($0.71 \times 0.71 \text{ cm}^2$, $0.50 \text{ cm}^2$). Each curve in Figure 3 represents one sample tested at a certain liquid pressure, and each data point in the curve represents values averaged over the time of one steady state measurement (at least 30 seconds). Figure 4 shows the high-speed camera images of the boiling surface at different heat fluxes for a specific test with the liquid pressure of 760 torr.

The boiling curves display a similar shape under different liquid pressure. At the beginning of the experiment with low heat flux, there was no bubble nucleation on top of the membrane. Water flooded on top of the membrane and formed a liquid layer, while the excess amount of liquid overflowed and dropped to the bottom of the vapor chamber. The liquid formed a thick puddle (>1 mm thick) on top of the membrane and the heat transfer mechanism was simply single phase convection (see Figure 4a). Bubble nucleation started after the wall superheat exceeded ~5 °C, where the bubbles burst and the liquid splashed inside the chamber (as seen in Figure 4b), and the heat flux was dissipated by both sensible and latent heat of the liquid. Since the heat flux causing the bubble nucleation (~5 W/cm$^2$) is usually orders of magnitude smaller than CHF in our experiments, this onset of nucleate boiling was not shown in the graph in Figure
3. With relatively low heat fluxes, the boiling curves have a small slope and the boiling behavior is in the nucleate boiling regime and is well captured by the Rohsenow correlation\textsuperscript{26}, as shown in Figure 3. This behavior can be attributed to the thick liquid layer on top of the membrane acting as a liquid pool, as in the case of pool boiling.

As the heat flux kept increasing, the boiling curves deviated from Rohsenow correlation and started to show a gradually larger slope, and, after a certain heat flux, an even negative slope. This behavior, a clear departure from the pool boiling as described by the Rohsenow correlation, can be attributed to the thin liquid film formation on the heater. When the film thickness is smaller than the bubble departure diameter in pool boiling (mm-scale), the boiling behavior would deviate from pool boiling.

The increasing and the eventual negative slopes in the boiling curves at the high heat flux are the hallmarks of the thin film boiling regime. The behavior can be attributed to the following factors: with increasing heat flux, as more liquid was vaporized while the mass flux was constant with a fixed pressure difference between liquid and vapor chambers ($\Delta P = P_l - P_v$), the mass of excess amount of liquid that remained on top of the membrane decreased, which yielded a decreasing liquid layer thickness. In fact, at higher $\Delta P$, the boiling curves deviated from Rohsenow correlation at a higher heat flux and wall superheat, which can be attributed to higher liquid mass flux delaying the onset of the thin film boiling regime. Previous work\textsuperscript{27} with Si micropillars also showed very similar thin film boiling behavior with high heat flux, in which the liquid film thickness was mainly determined by the pillar height.

The negative slopes in the boiling curves, that is, a decrease in wall superheat with
increasing heat flux at high fluxes, can also be understood from the thin film geometry. Similar phenomena have been observed in the literature and were usually due to the hysteresis associated with the activation of nucleation sites, and therefore were usually observed at the onset of the nucleate boiling. However, this negative slope, termed “boiling inversion” in Kandlikar’s work\textsuperscript{28}, could also result from enhanced macroconvection\textsuperscript{29}. In our study, due to the unique thin liquid film configuration, three possible mechanisms can explain these negative slopes, namely, the enhanced conduction inside the liquid layer, the promoted bubble departure, and the separate liquid-vapor pathways.

Thinner liquid layer at higher heat flux reduces the conduction thermal resistance inside the liquid layer and consequently provides inertia-controlled bubble growth. We hereby analyze an exemplary boiling curve with $\Delta P = 1259.5$ torr (the magenta curve in Figure 3) as follows. Hsu\textsuperscript{30} suggested the criteria for the incipience of nucleation for pool boiling as $\frac{12.8\sigma T_{\text{sat}}(P_t)}{\rho_v h_{fg} \delta_t \theta_w} = 1$, where $\sigma$, $\rho_v$, $h_{fg}$ are the surface tension, vapor density, and latent heat of water, respectively. In this particular test, bubbles were observed at a wall superheat of 16.2 °C (which is the first point recorded on this curve). Therefore, the incipience of nucleation occurred before the wall superheat $\theta_w$ reached 16.2 °C, which yields a thermal boundary layer thickness $\delta_t$ larger than 395 µm. The liquid layer thickness would decrease from >1 mm at zero heat flux to zero at CHF (since the layer dries out at CHF), which means that at certain heat flux, the liquid layer thickness will be smaller than this thermal boundary layer thickness. It was reported\textsuperscript{31} that a liquid layer as thin as ~105 µm could be obtained in a boiling thin liquid film, which is smaller than the thermal boundary layer thickness calculated above. Therefore, at sufficiently high heat
flux, the liquid layer on top of the membrane will be superheated and the temperature of the whole liquid layer will be close to the wall temperature as depicted in Figure 1b. The small thickness and the superheating of the liquid pool resulted in the enhanced conduction inside the liquid layer and therefore the bubble growth of the boiling process was entirely in the inertia-controlled regime, in which all of the liquid-vapor interface area was efficiently evaporating and the bubble growth was fast for the entire growth period.

The second possible reason for the reduced overall thermal resistance is the promoted bubble departure in the thin liquid layer. For an extensive liquid pool, the bubble departure diameter $d_d$ is usually comparable with the capillary length $l_c$ given by

$$l_c = \sqrt{\frac{\sigma}{g(\rho_l - \rho_v)}}$$

Given water properties at 20 °C, $l_c \approx 2.7$ mm is at least one order of magnitude larger than the liquid pool thickness at high heat fluxes. Therefore, the bubbles in the thin liquid film would depart before growing to the size of $l_c$; hence, the real bubble departure diameter would be close to the liquid layer thickness. Although it is difficult to obtain a clear bubble boundary from the images at high heat flux, it can still be seen that the relative bubble size in Figure 4(d-f) (in the high heat flux regime) is much smaller than both the characteristic length of $\sim 2.7$ mm and the bubble size at low to moderate heat flux in Figure 4b and c. This indicates that the bubble departure will happen much more frequently than that in normal pool boiling with the same superheat, which can also be responsible for the greatly enhanced heat flux beyond the Rohsenow regime.

Another possible mechanism responsible for the heat transfer enhancement is the efficient liquid replenishment. Since the heat flux was supplied to the solid wall of the membrane, the nucleation will more likely to occur at the top of the solid walls and liquid is replenished after...
bubble departure. As shown in Figure 1b, we hypothesize that bubbles depart vertically upward while liquid was supplied to the nucleation sites laterally from the pores without interference, providing enhanced macro-convection heat transfer\textsuperscript{29}, which has been shown previously to cause boiling inversion\textsuperscript{25, 28, 29}. The separate liquid-vapor pathways provided by the nanoporous configuration can also partly explain the heat transfer enhancement. Also, the lateral length-scale needed for liquid to travel in order to replenish the nucleation sites is on the order of several hundred nanometers (determined by the pitch size between pores) and is much shorter compared with mm-scale (determined by bubble size) as in the case of pool boiling.

From Figure 3, it can be seen that the boiling heat flux of the nanoporous configuration in this work is clearly dependent on the pressure difference ($\Delta P$) between the liquid chamber ($P_l$) and the vapor chamber ($P_v$). The CHF of nanoporous surface increases with increasing $\Delta P$, which can be explained by the limiting mechanism for CHF in the current configuration. In some of the boiling experiments on micro- or nanostructures, the capillary limit is the ultimate limit for the CHF condition\textsuperscript{32-36}. Liquid-vapor interfaces confined in these structures generate large capillary force which drives the liquid to the phase-change sites and the CHF is achieved when the capillary driving pressure is balanced by the viscous drag of liquid transport. However, in our current configuration, the membrane was always covered by the liquid, and there was no meniscus liquid-vapor interface inside the nanopores of the AAO membrane. This is clearly shown in Figure 5, which depicts the CHF vs. $\Delta P$ with the linear fitting of the plot passing through the origin, meaning a zero heat flux (and mass flux) corresponding to no external liquid pumping pressure. Therefore, the driving force for liquid transport across the membrane comes
entirely from the external pressure difference across the membrane, i.e., $\Delta P$ between the liquid chamber and the vapor chamber (the hydraulic resistance in the tubing is negligible). This pressure difference will provide an almost constant liquid mass flux inside the nanopores for each individual experiment with fixed $\Delta P$ (assuming the liquid properties are insensitive to temperature and pressure). CHF is achieved when the mass flux is completely vaporized and further increase of heat flux would result in membrane dry out. Therefore, the increased $\Delta P$ in this configuration is thus similar to increased mass flux in a flow boiling device.

The maximum heat flux that can be dissipated before membrane dry out can be expressed as

$$q''_t = \dot{m}_{tot}(h_{fg} + c_p\Delta T)/A$$

where $\dot{m}_{tot}$ is the total mass flux provided by the pressure difference. According to the laminar viscous flow theory, $\dot{m}_{tot}$ is proportional to $\Delta P$, indicating that the total heat flux $q''_t$ is also proportional to $\Delta P$, since water density and latent heat do not vary too much with temperature and the sensible heat is an order of magnitude smaller than latent heat given the small temperature rise in the experiments. Figure 5 clearly shows that the experimental CHFs are linearly related to $\Delta P$ and match well with the theoretical results with wall superheat at 60 °C (see Supporting Information for details on the modeling).

The maximum CHF of 1230.7 W/cm² reported here is among the highest numbers for boiling heat transfer. It should be noted that this CHF has so far only been limited by mechanical strength of the membrane. By increasing the pressure difference to provide larger liquid mass flux or by decreasing the membrane thickness to reduce the liquid flow resistance, higher CHF and HTC can be achieved. However, both of these approaches compromised the mechanical strength of the devices and have led to fracturing of the membranes. Better membrane design
with support structure or using other materials with better strength should be used for future pursuit of higher heat flux.

Although the liquid supply for vaporization in the present thin film boiling configuration is similar to traditional flow boiling, it does not experience the flow instabilities commonly seen in flow boiling. During the experiments, the dynamic temperature profiles at each heat flux step was recorded and averaged. Figure 6 shows the temperature and heat flux profiles of one typical experiment (CHF=1230 W/cm² with liquid pressure at 1398 torr). As opposed to flow boiling where dramatic temperature fluctuation exists, the current configuration showed stable wall temperature at all the range of heat fluxes with a small standard deviation of 0.19, 0.19, and 0.27 °C for heat fluxes of 12, 707, and 1230 W/cm², respectively. At low heat flux, similar to homogeneous nucleate boiling, temperature was stable because the bubbles at the local nucleation sites only occupy a small fraction of the entire area. The stability of temperature at higher heat flux can be attributed to the way liquid was supplied as well as the small thickness of the liquid layer. In flow boiling, due to the large volume expansion of liquid-vapor phase change as well as the compressibility of vapor in the confined channel, flow instabilities such as Ledinegg instability and pressure drop instability tend to happen. In our current thin film boiling, liquid was supplied from the cross-plane direction through nanoscale-confined pores, which suppressed the cavitation and bubble formation inside the pores. The phase change phenomenon happened on top of the membrane in a large vapor space, and therefore, flow instabilities can be eliminated. Furthermore, unlike flow boiling where the liquid mass flux decreases along the channel due to vaporization, in this configuration, the liquid mass flux at all
locations of the heater surface was nearly the same. The thin liquid layer can also constrain the bubbles to a small diameter and prevent vapor accumulation. Therefore, the lateral coalescence of bubbles and the resulting formation of vapor blanket can be eliminated until the heat flux approaches CHF where vapor volume fraction is sufficiently high to form a local hotspot and cause dry-out.

The heat flux observed in our study, up to ~1.23 kW/cm\(^2\), is at least 10 times higher than previous work by Wang and coworkers\textsuperscript{40,41} on similar setups using AAO membranes. In Refs.\textsuperscript{40,41}, after the top liquid layer dried out and all the liquid covering the membrane was vaporized, thin film evaporation from the nanopores will continue dissipating higher heat flux. Supposedly, after heat flux reaches a certain value such that the liquid covering the membrane is completely dried out, the liquid-vapor interface will recede inside the nanopores and form a meniscus shape, and consequently provide extra driving force for liquid supply by the meniscus-generated capillary pressure, as clearly observed in Refs.\textsuperscript{40,41}. However, in our experiments, from visual observation (Figure 4f) as well as the dynamic temperature measurement in the experiment (Figure 6), the wall temperature kept increasing and could not reach a steady state as soon as a dry spot appeared on top of the membrane (Figure 6 and Supporting Information Figure S6); this overheating eventually caused membrane failure and the CHF. Figure 4f shows a typical image right before the CHF, which indicated that the central area of the membrane was dried out. Therefore, we infer that the evaporation regime with receding meniscus inside the pores could not be reached under our experimental conditions. This is further supported by the fact that the linear fitting of the CHF vs \(\Delta P\) in Figure 5 has zero intercept at zero pressure difference,
indicating the absence of the capillary liquid pumping.

Combining our observation and those from Wang and coworkers\textsuperscript{40, 41}, we suggest that the condition to reach the capillary-driven evaporation regime (with menisci residing inside the nanopores) depends on the heat flux prior to the dry out of the liquid layer covering the membrane. If the heat flux prior to the dry out is lower than what could be achieved in the capillary-driven evaporation inside the nanopores, then, with increasing heat flux, the phase change will transition to the evaporation regime after the dry out, as observed by Wang and coworkers\textsuperscript{40, 41}, where the heat flux for this transition was below 30 W/cm\textsuperscript{2} due to the much lower permeability of the AAO membranes they used (smaller pore size and lower porosity). In our experiments, the CHF in the thin film boiling regime (> 1 kW/cm\textsuperscript{2}) is likely already higher than the heat flux attainable from the evaporation inside the nanopores. Therefore, a stable evaporation regime could not be reached.

The remaining question is, of course, why the heat flux from the evaporation inside the nanopores is not higher (say, > 1 kW/cm\textsuperscript{2}). We hypothesize that the most probable reason is the vapor diffusion limit of the evaporation inside pores. The evaporation heat flux from nano-pores could be governed by three transport resistances: the hydraulic resistance of liquid transport, the kinetic limited interfacial resistance of evaporation, and the vapor diffusion resistance from interface to the far field in the vapor space. We can first eliminate the hydraulic resistance limit because the capillary force would have provided extra driving pressure of several hundred kPa, which would have led to a heat flux much higher than 1 kW/cm\textsuperscript{2}, as we can infer from the heat flux vs. \(\Delta P\) plot in Figure 5. Second, we can also estimate that the kinetic limit would have been
much higher. Previous modeling work\textsuperscript{42, 43} has shown that the kinetic limited heat flux of evaporation from nanopores can reach as high as several kW/cm\textsuperscript{2}. Recently, Duan and coworkers\textsuperscript{44} experimentally measured the kinetic limit of water evaporation inside nanochannels and found that the kinetic limited evaporation heat flux was even an order of magnitude larger than theoretical prediction. Therefore, the heat flux at the moment of liquid layer dry-out in our experiments was still lower than the kinetic limit. Therefore, we hypothesize that the vapor diffusion limit would have prevented the liquid from receding into the nanopores and forming evaporating menisci. As described in Ref.\textsuperscript{44}, the evaporation flux from each channel would be suppressed when the nanochannel was adjacent to other channels, indicating the vapor removal being the limit for evaporation. In our experimental configuration, the nanopores are closely packed with a pore-to-pore distance of \textasciitilde100 nm, which is highly possible to induce a very large vapor advection/diffusion resistance to limit the evaporation heat flux. In our experiments, as the maximum evaporation heat flux limited by vapor diffusion was smaller than the maximum heat flux attainable in the thin-film boiling regime when liquid-layer-dryout heat flux, no thin film evaporation regime could be stably reached.

Returning to our initial question regarding the upper limit of heat flux from a planar surface, it would be interesting to compare our results to the theoretical limit in phase change heat transfer. Although bubbles are present in the thin film boiling regime, the ultimate upper limit for the heat flux is essentially the same as the maximum interfacial heat flux on a planar surface, which is achieved when all of the molecules at the liquid-vapor interface would escape to the vapor phase with certain speed (usually considered on the order of the speed of sound\textsuperscript{11, 45}) and
without returning to the liquid, i.e., $q''_{max} = \rho_v c h_{fg}$ where $c$ is the average vapor speed. However, this simple equation only accounts for the mass conservation, and does not represent the non-equilibrium behavior in the Knudsen layer during an intensive evaporation. Here we employed an approximate method from the Boltzmann transport equation with consideration of conservation of mass, momentum and energy to describe the interfacial flux of intensive evaporation\textsuperscript{46}. The calculated interfacial flux for a liquid layer at 70 °C (corresponding to the superheat of 50°C at the CHF condition in Figure 3) evaporating to the vapor space at 20 °C is close to 5 kW/cm\textsuperscript{2} (see Supporting Information S8). Our experimental CHF is only within a factor of 4 of this limit. Moreover, this CHF can be increased by improving the liquid pumping pressure in more mechanically robust membranes.

In summary, we realized a new “thin film boiling” mechanism using Nanoporous membranes to achieve the highest CHF of boiling heat transfer on a planar surface. By controlling liquid pressure while maintaining a constant vapor pressure, different mass flux across the membrane, and consequently different CHF values, can be provided. The highest CHF (1230 W/cm\textsuperscript{2}), achieved at a liquid pumping pressure of ~1398 torr, is within a factor of four of the upper limit of CHF, and was only limited by the mechanical strength of the membrane. Increasing and eventually negative slopes were observed in the boiling curves due to the thin liquid film geometry. The enhanced boiling performance was attributed to the separate liquid-vapor pathways, the thin liquid layer promoted bubble departure, and the reduced conductive thermal resistance. This study also provides a new approach for achieving ultrahigh heat flux in boiling heat transfer for thermal management of high power devices.
ASSOCIATED CONTENT

Supporting Information. Membrane characterization; TCR calibration of the Pt layer; conduction heat loss; error from water electrolysis; error analysis; experimental procedure; theoretical CHF modelling; upper limit of the CHF. This material is available free of charge via the Internet at http://pubs.acs.org.

Author contributions. Q.W. and R.C. conceived the research idea, designed the experiments, and wrote the manuscript. Q.W. carried out the experiments and the theoretical modeling.

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Notes. The authors declare no competing financial interest.
Figure 1. Schematics of the bubble growth behavior in (a) pool boiling and (b) thin film boiling.

Different from pool boiling, when the liquid film thickness is smaller than the thermal boundary layer thickness in thin film boiling, the bubble growth is in the inertia-controlled regime where all the interface area participates in efficient evaporation. The bubble departure diameter is limited by the liquid film thickness. The liquid replenishment from the pores reduces the lateral length for liquid transport.
Figure 2. (a) and (b): SEM images of the AAO membrane, (a) Top view; (b) cross-sectional view. Scale bar: (a) 1 µm (inset: 200 nm); (b) 50 µm (inset: 500 nm). (c) Schematic and photo (zoomed-in bottom) of the sample assembly for the experiment.
Figure 3. Boiling curves under different liquid pressures. The vapor pressure was maintained at 17.5 torr (20°C saturation temperature). The arrows indicate the CHF for each curve. The highest CHF of 1230.7 W/cm² was achieved with liquid pressure at 1398 torr.
Figure 4. High speed images of the boiling surface at different heat fluxes under 760 torr liquid pressure. (a) 0 W/cm$^2$, thick liquid puddle formed on top of the membrane, the white part was the reflection of light from the large liquid puddle; (b) 3.7 W/cm$^2$, bubble nucleation started; (c) 55.2 W/cm$^2$, a clear bubble base shows mm-scale bubble size; (d) 337.5 W/cm$^2$, bubbles were smaller than Rohsenow regime; (e) 571 W/cm$^2$, much smaller bubbles formed over the entire area; (f) 700 W/cm$^2$ (near CHF), center dry out occurred on top of the membrane, while the surrounding area still showed small bubbles.
Figure 5. Experimental and theoretical CHFs vs. pressure difference. The experimental CHFs and modelling for theoretical CHFs with 60±20 °C superheat agree well. The linear extrapolation of the curve has a zero intercept at zero pressure difference, meaning no capillary pumping effect. The theoretical CHF model used Hagen-Poiseuille equation through circular pores to calculate the liquid mass flux and assumed all of the liquid was vaporized (see Supporting Information S7 for details).
Figure 6. Dynamic temperature and heat flux profiles. (a) Temperature as a function of time. (b) Heat flux as a function of time. Data represents the experiment with 1398 torr liquid pressure (CHF at 1230 W/cm²).
REFERENCES

44. Li, Y.; Alibakhshi, M. A.; Zhao, Y.; Duan, C. *Nano Letters* **2017**.