Minimum film-boiling quench temperature increase by CuO porous-microstructure coating

Jun-young Kang,1 Qi Cheol Lee,2 Massoud Kaviany,1,3 Hyun Sun Park,1 Kiyofumi Moriyama,1 and Moo Hwan Kim1(a)

1Division of Advanced Nuclear Engineering, POSTECH, Pohang 790-784, South Korea
2Department of Mechanical Engineering, POSTECH, Pohang 790-784, South Korea
3Department of Mechanical Engineering, University of Michigan, Ann Arbor, Michigan 48109, USA

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Increase in the minimum film-boiling quench temperature, $T_{\text{MFB}}$, is achieved with micromstructured CuO particles, and attributed to local cooling (fin effect) by the microstructure causing liquid–solid contact. A periodic structure is obtained using electrochemical deposition of 1 µm diameter particles on brass sphere diameter 15 mm forming unit-cell porous cones of average height $L = 100 \mu m$ and base diameter $D = 20 \mu m$. Fin analysis predicts the cone tip cooling to the homogeneous nucleation temperature of water ($\sim 330^\circ C$), while the base temperature is at 600 $^\circ C$. This causes liquid–solid contact during quenching, and analysis suggests the fin effective thermal conductivity $\langle k \rangle$ and fin characteristic length $L_\text{f}/D$ are key to this liquid–solid contact that influences $T_{\text{MFB}}$. Published by AIP Publishing. [http://dx.doi.org/10.1063/1.4974923]

The quench-boiling cooling is hindered by the vapor-film insulation in the film-boiling regime,1 and the minimum film-boiling quench temperature $T_{\text{MFB}}$ for stable film is important in sudden cooling of high-temperature systems.2 The $T_{\text{MFB}}$ is strongly influenced by surface conditions including any microstructure,3–8 such as porous coatings using micron or sub-micron particles,9–12 and this has been associated with causing liquid–solid contact during boiling.13 Yet, the role of microstructures in allowing for the liquid–solid contact and the control of structure have not been explored. Control of $T_{\text{MFB}}$ is one of the unknowns in boiling phenomena, i.e., “what is the extent and character of liquid-solid contact in film and film-transition boiling?”.14 Strategy to promote the liquid-solid contact by this surface microstructure leads to $T_{\text{MFB}}$ increase and contributes to the development of ultra-fast quench system at nuclear power plants, cryogenics, and metallurgy. In this letter, we report on the quench $T_{\text{MFB}}$ increase using CuO porous microstructure coating (MPC), and explain the role of the microstructure properties. The electrochemical deposition (ECD) enabled the formation of periodic porous cones with an average particle diameter $d_p = 1 \mu m$, average height $L = 100 \mu m$, and base diameter $D = 20 \mu m$. We explain that quenching results from local cooling, caused by the microstructures, in accordance with the fin theory, and suggest physical parameters controlling the liquid–solid contact.

The quenching experiments are conducted with atmospheric saturated water quenching a sphere (brass, $d_{\text{sphere}} = 15 \text{ mm}$), as shown in Figs. 1(a), and S1 [supplementary material]. The test surfaces were cleaned with acetone, ethanol, and water, and polished with abrasive paper (CW2000-2C; Daesung). We tested three surfaces: the plain surface (PS), and two coated surfaces, the MPC, and a nanostructured coating (NC). The MPC and NC surfaces were fabricated by heat-enhanced ECD which forms CuO particles,15–17 and the method of Xiao et al.17 [detail in supplementary material S2]. The surface morphology, chemical composition, and surface wettability (contact angle $\theta_c$) were evaluated using field-emission scanning electron microscopy (FE-SEM) with energy dispersive spectrometry (EDS) (JSM7401F; JEOL), X-ray diffraction (XRD) (D/MAX-2500/PC; Rigaku), and a contact angle measurement system (SmartDrop; Femtobaf), as shown in Figs. 1(b)–1(f) and supplementary material S3. The MPC surface morphology is formed by interaction between two reaction products (H$_2$(g) and CuO(s)) during the ECD process; CuO microparticles cannot uniformly coat the quench sphere surface, due to generation of the H$_2$ gas and, the CuO microparticles for periodic porous cones. The chemical composition of the MPC and NC was identified as CuO through EDS analysis: Cu(≈55%) and O(≈45%). The geometric parameters ($L$ and $D$) in surface structures and $\theta_c$ of each test surface are summarized in Table SI (supplementary material S3).

The quench time depends on the surface conditions, and is 57 s for the PS, 50 s for the NC, and 7 s for the MPC, as shown in Fig. 2(a). The temperature history presented as $dT/dt$ before $T_{\text{MFB}}$ is characteristic of film-boiling by heat conduction and radiation across the vapor film (8.3 $C/s$ for PS and NC in the range 400 $< T < 550^\circ C$). The $T_{\text{MFB}}$ of NC (300 $^\circ C$) was higher than that of PS (250 $^\circ C$), owing to the improvement in surface wettability. However, the MPC surface has a $T_{\text{MFB}} > 600^\circ C$ and its large $dT/dt$ (60 $C/s$ in the range 400 $< T < 550^\circ C$) cannot be explained by vapor conduction film boiling, $q = k_v(T_w - T_{\text{sat}})/\delta_v$, where $q$, $k_v$, $T_w$, $T_{\text{sat}}$, and $\delta_v$ are the heat flux, vapor thermal conductivity, wall temperature, saturation temperature, and vapor film thickness. This result is consistent with that shown by the high-speed snapshot (Figs. 2(b) and 2(c)) (Multimedia view) captured during the quenching, and the boiling curve (Fig. S4, supplementary material). The liquid–vapor interface was initially perturbed when the PS sphere was immersed in the
saturated water, but the stability was recovered, and the film boiling was maintained until the temperature had reached $T_{\text{MFB}}$. The NC sphere behaved in a manner similar to that of the PS, with the collapse of the film occurring quicker than the PS sphere. However, the film boiling on the MPC sphere was unstable, with an explosive evaporation. The liquid–vapor interface was unstable until quenching was complete. This observation implies that the heat transfer mechanism for the MPC differs significantly from the other surface conditions, due to liquid–solid contact throughout quenching.

Several mechanisms cause the liquid–solid contact related to $T_{\text{MFB}}$. First, it has been suggested that the vapor flowing into the porous medium can be absorbed, resulting in $T_{\text{MFB}}$ increase.\textsuperscript{18} The permeability $K$ of the porous microstructure from the Carman–Kozeny relation, $K = \phi^3 d_p^2 / [180 (1-\phi)^2]$, for the MPC sample with porosity $\phi = 0.5$, and $d_p = 1\, \mu m$ gives $K \sim 2.7 \times 10^{-15} \, \text{m}^2$ (S5, supplementary material), which is too small for a significant permeation. Secondly, the destabilization of the liquid–vapor interface can be attributed to the hydrophilic nature of the metal-oxide microparticles,\textsuperscript{19} because a liquid can easily form menisci at the intermittent liquid–solid contact.\textsuperscript{20,21} However, an increase in $T_{\text{MFB}}$ does not solely result from hydrophilicity, as was demonstrated by the NC surface, which is in agreement with the observations of Kang et al.\textsuperscript{6} and Fan et al.\textsuperscript{22,23} as $T_{\text{MFB}}$ increase on super-hydrophilic surfaces. Lastly, while a high $L/d_v$ ratio allows for the liquid–solid contact, it is not a general rule, as reported by Kozlov and Keßler;\textsuperscript{7} an increase in $T_{\text{MFB}}$ is not solely determined by $L/d_v$. Therefore, describing the liquid–solid contact for the MPC sample using the mechanisms discussed above is insufficient. It is known that $T_w$ should be smaller than the homogeneous nucleation temperature, $T_{\text{hn}}$, of water ($\sim 330^\circ\text{C}$), for maintaining the liquid–solid contact during film boiling,\textsuperscript{24} because the liquid phase above $T_{\text{hn}}$ must approach the superheat limit. The liquid can make contact the quench surface if it has a local cold region with a temperature that is close to $T_{\text{hn}}$. This is similar to the effects of an insulation layer during quenching.\textsuperscript{25,26} Therefore, the temperature gradient of the microstructures needs a close evaluation using the fin theory to determine the possibility of liquid–solid contact.

Considering the liquid–solid contact time $\tau_c \sim 10^{-1} \, \text{s}$,\textsuperscript{27,28} the transient cooling of the microstructures is
simplified to quasi-steady state, one-dimensional fin analysis due to a large axial fin Fourier number $F_{oL} = \alpha \tau_c L^2$, and small radial fin Biot number $B_{iD} = hD/(k)$ for the MPC surface ($F_{oL} > 1$, and $B_{iD} \ll 10^{-3}$).\textsuperscript{29} where $\alpha$, $h$, and $L$ are the fin thermal diffusivity, heat transfer coefficient, and effective thermal conductivity of microstructures. The fin calculation is based on the Murray–Gardner assumptions.\textsuperscript{30} The ambient temperature $T_{\text{amb}}$ was assumed to be the film temperature $T_i = (T_w + T_{\text{sat}})/2$. The geometry of the microstructures is conical spine, as shown in Fig. 3(a), and $\langle k \rangle$ was calculated from the relation $\langle k \rangle = \varphi k_s + (1 - \varphi) k_{\text{CuO}} \approx 0.5$ W/(m K) when $\varphi = 0.5$, $k_s = 0.05$ W/(m K), and thermal conductivity of cupric oxide $k_{\text{CuO}} = 1$ W/(m K).\textsuperscript{31} The radiation heat flux $q_{\text{rad}}$ is negligible in the temperature range $q_{\text{rad}} = \sigma_{\text{SB}} (T^4 - T_{\text{sat}}^4)/(1/(k_{\text{CuO}} + (1/\varepsilon_t) - 1) \approx 30$ kW/m$^2$, where the Stefan–Boltzmann constant is $\sigma_{\text{SB}} = 5.67 \times 10^{-8}$ W/(m$^2$ K$^4$), the emissivity of cupric oxide and liquid are $\varepsilon_{\text{CuO}} = 1$ and $\varepsilon_t = 1$, and $T = 600$ °C.\textsuperscript{32} The $h$ is calculated using the lumped parameter method, due to the small sphere Biot number; $B_{iD} = hD/k_{\text{sphere}}$ < 0.1 where $d_{\text{sphere}}$ and $k_{\text{sphere}}$ are the fin-tip and fin-base temperature, respectively. The temperature difference in microstructures (between fin-tip and fin-base) is strongly dependent on the dimensionless length $mL = [2hL^4/(\langle k \rangle D)]^{1/2}$, i.e. 

$$
\theta^* = \left(1/\zeta^*\right)^{1/2}I_1 \left[2^{3/2}(mL\zeta^*)^{1/2}\right]/I_1 \left(2^{1/2}mL\right),
$$

(1)

where $\theta^*$, $\zeta^*$, and $I_1$ are the dimensionless temperature difference ($T_{x = 0} - T_{\text{amb}}$)($T_{x = 1} - T_{\text{amb}}$), dimensionless axial position ($x/L$), and the modified Bessel function of the first kind of order one, respectively (supplementary material, S6). $T_{x = 0}$ and $T_{x = 1}$ are fin-tip and fin-base temperature, respectively. As $mL$ increases, $\theta^*$ decreases significantly, as shown in Fig. 3(b). For example, $\theta^*$ at $x = 0$ was less than 0.1 for the MPC ($mL \approx 1.3$), and its tip temperature is close to $T_{\text{amb}}$ of water. It can be assumed that microstructures at low $mL$ did not cause the local temperature decrease during quenching; $\theta^*$ at $x = 0$ is 1 under $mL = 0.06$, where $h = 800$ W/(m$^2$ K), $L = 100$ µm, $D = 20$ µm, and $k_{\text{basal}} = 100$ W/(m K). This result is identical to that of Kim et al.\textsuperscript{34} the increase in $L_{\text{MFB}}$ varies depending on the thermal property of the microstructures (SiO$_2$, Al$_2$O$_3$, and diamond). A low fin efficiency $\eta = 2^{1/2}I_2(2^{1/2}mL)/[mL(2^{1/2}mL)]$ implies an increase in surface area has little effect on the increase in heat transfer ($\eta < 0.6$ for MPC, and $I_2$ is the modified Bessel function of the first kind of order two). Therefore, porous microstructures can promote liquid–solid contact, even though the quench temperature is 600°C, and this prevents stable film boiling.

To illustrate the local effect of porous microstructures, note the ratio of the spot area $A_{\text{spot}}$ and the total area of the sphere $A_{\text{sphere}}$ is about $10^{-3}$, as shown in Figs. 4(a) and 4(b). The liquid–solid contact of the microstructures produced liquid–vapor interface with different dynamics (Fig. 4(c)) (multimedia view). The observed variation of the liquid–vapor interface is explained through analysis of the liquid–solid contact, and this phenomena maintains until the quench is over; clear difference in this phenomena is available in supplementary material 7 (image processing conversion). The large $mL$ (through $\langle k \rangle$ and fin characteristic length $L^2/D$) of CuO porous microstructures improves liquid contact with solid, increasing $L_{\text{MFB}}$. Small $\langle k \rangle$ and large $L^2/D$ cause significant fin tip-cooling, and this is independent of the surface super-hydrophilicity. Microstructures with large $mL$ cause accelerated cooling in high-temperature quenching and the ECD method is promising for such surface modifications.

See supplementary material (S1) for the apparatus of quenching of high temperature sphere, (S2) for coating preparation on quench sphere, (S3) for surface parameters in quench spheres, (S4) for boiling curve, (S5) for vapor absorption in permeable surface, (S6) for fin theory of microstructures, and multimedia video to illustrate the liquid-vapor interface in quench are in (S7).
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