带有超疏水点阵的微细铜丝上的核态沸腾

王新巍, 王昊*

北京大学工学院, 北京 100871

* 联系人, E-mail: hwang@coe.pku.edu.cn

2019-09-12 收稿, 2019-10-16 修回, 2019-10-16 接受, 2019-12-25 网络版发表

国家自然科学基金(51622601)资助

摘要 表面亲疏水特性对沸腾有着重要影响. 本文在直径为150 μm 的铜丝上修饰了宽度为100 μm 、间距为3 mm的超疏水点阵. 借助微细丝较好的观测特性, 对疏水点阵上的沸腾过程, 尤其是气泡脱离及相互合并过程进行了细致考察. 研究了单个疏水点大小对起泡的影响, 并对带有疏水点阵的铜丝上的沸腾换热模型进行了初步考察. 超疏水点阵可以显著提高核态沸腾换热效率, 初步认为其主要原因在于点阵使得起泡更加容易, 且沿加热丝更加均匀, 但最优的核化点密度及其与铜丝的固体热性能的关系还有待进一步研究.

关键词 核态沸腾, 亲水, 超疏水, 气泡, 换热强化

固体表面的物理化学性质(包括粗糙度、亲疏水特性)对沸腾传热有着重要影响^[1]. 亲水表面有助于气泡底部液体浸润, 从而提高临界热流密度(critical heat flux, CHF)^[2]. 疏水表面富含气泡, 有助于降低起泡温度^[3], 但容易造成干斑, 以致降低高热流下的换热效率以及临界热流密度^[4]. 近年来, 多项研究尝试在亲水基底上加工疏水点阵, 以结合亲、疏水表面的优点^[5~8]. 研究^[5,6,9]发现, 这类组合型表面可以显著提升核态沸腾换热效率. 少量研究对这类组合表面的起泡和脱离行为进行了观察^[5,8], 但是由于这类表面通常都是平板表面, 给细致观测带来难度. 换热效率提升的机制也未能全面了解. 本文利用带有超疏水点阵的微细铜丝作为实验对象, 实现了沸腾行为的有效观测, 并对相关强化机理做出推测.

1 实验设置

将直径为150 μm 的细铜丝用作沸腾表面. 首先, 在铜丝表面均匀地涂覆光刻胶(BP212, 北京微电子研究

所研制), 放在90°C的热板上烘烤10 min, 使光刻胶充分固化. 其次, 将带有预先设计好图案的玻璃光刻掩模板盖压在铜丝上, 并通过紫外线照射, 使被照射到的光刻胶产生光化学反应. 将铜丝放在80°C的热板上烘烤5 min, 增强光刻胶与铜丝表面的接触. 用显影液将被紫外线照射过的光刻胶洗去, 并用去离子水将铜丝表面的显影液冲洗干净, 最终在铜丝上形成带有一系列通孔的光刻胶虚拟掩模. 然后, 通过非电解电镀沉淀法^[10]实现超疏水点阵. 具体地, 将经影印石版术处理后的铜丝浸入浓度为0.010 mol/L的硝酸银溶液, 使得硝酸银溶液进入光刻胶虚拟掩模的通孔阵列与铜丝表面产生银镜反应, 反应20 s后在铜表面形成致密的纯银薄层点阵. 之后将铜丝浸入1 mmol/L的HDFT溶液, 反应5 min, 使纯银薄层与HDFT充分反应得到超疏水点阵. 用质量分数为5%的NaOH溶液将点阵周围的光刻胶清洗掉, 最终在铜丝上形成微尺度的超疏水点阵. 如图1左下所示, 本研究主要使用带有宽度为100 μm 超疏水点, 点间距为3 mm的铜丝作为加热丝进行实验. 经测

引用格式: 王新巍, 王昊. 带有超疏水点阵的微细铜丝上的核态沸腾. 科学通报, 2020, 65: 1734~1740

Wang X W, Wang H. Nucleate boiling on thin copper wires with micropatterned superhydrophobic and hydrophilic areas (in Chinese). Chin Sci Bull, 2020, 65: 1734~1740, doi: [10.1360/TB-2019-0518](https://doi.org/10.1360/TB-2019-0518)

图1 (网络版彩色)实验设置以及带疏水点阵的铜丝

Figure 1 (Color online) The experimental setup and the copper wire patterned with superhydrophobic areas

量, 超疏水点的接触角大于 170° .

实验中使用的沸腾发生装置主要由两端连接到铜电极上的铜丝、不锈钢主容器、直流电源和压力传感器组成, 如图1所示. 铜丝的长度为85 mm、直径为 $150\ \mu\text{m}$, 表面粗糙度RZ约为600 nm, 纯度为99.95%. 通过测量铜丝两端电压以及通过的直流电流, 可以计算其电阻和发热功率. 利用实验前标定好的铜丝电阻-温度换算关系, 求得加热丝的平均温度, 计算出壁面过热度.

容器内主液区的温度由3支型号相同的PT100铂电阻同时进行测量, 每支铂电阻的精度均为 0.1°C . 3支铂电阻的水平位置不同, 但高度基本相同, 均垂直距离加热丝2 mm. 实验过程中, 它们的读数相差不超过 0.2°C , 因此可取任意一支铂电阻的读数作为主液区的温度值. 为实现主液区温度控制, 在容器底部放置一个辅助加热棒, 温控器在收到铂电阻测得温度的反馈后, 对主液区工质进行温度调节. 容器上方安装的压力传感器能够对容器内的压力实时监测, 根据实测的压力值调节调压阀, 实现容器内压力的控制.

高速摄像系统主要由计算机、高速摄像头、体视显微镜、摄像照明系统组成. 高速摄像头为X-Motion, AOS Technology, 最大拍摄速度可达到30000帧/s, 实验中通常使用10000帧/s. 体视显微镜为SZ-CTV, OLYMPUS, 物镜最大放大倍数为11倍, 实验中通常使用5倍物镜对气泡进行观察. 将高速摄像头一端与体视显微镜

连接, 另一端通过专用网线与计算机相连, 组成成像系统. 摄像照明系统为冷光源Multi-Position, Nikon.

实验开始前, 先将工质煮沸1 h, 之后调整辅助加热器保持工质处于饱和状态. 然后铜丝先在低热流下沸腾约5 min, 以减小不凝结性气体的影响. 之后对铜丝施以特定电功率加热, 待实验现象稳定后记录气泡行为、加热丝电流、电压以及主液区温度数据. 同时将成像系统与冷光源分置于沸腾发生装置的两侧, 启动高速摄影系统, 调整焦距、光源等, 记录清晰的气泡图像.

2 结果与讨论

在正式开展疏水点阵实验之前, 首先直观对比亲、疏水表面沸腾的不同. 如图2所示, 在亲疏水分界线的左侧为全面覆盖的超疏水区域, 右侧为裸露铜丝区域(即亲水区域). 可以看出, 在相同热流密度情况下, 当右侧的铜表面上还处于离散气泡核态沸腾阶段时, 左侧的疏水区域上已经形成稳定的汽膜.

而在疏水点阵铜丝上, 如图3和4所示, 气泡行为被有效控制. 图3显示起泡及脱离的全过程. 可以看到, 起泡就在超疏水核化点上发生. 当气泡准备脱离时, 气泡根部出现一个颈部, 颈部缩小直至断裂, 使得气泡得以脱离. 少量可观测到的气体残余在核化点位置, 成为下一个气泡周期的起始.

随着热流密度的逐渐增加, 超疏水点阵铜丝上的沸

图2 超疏水表面与亲水表面沸腾现象对比. 左侧超疏水区域全面覆盖了超疏水涂层, 右侧为裸露铜丝表面

Figure 2 Comparison of boiling on uniform superhydrophobic coating and hydrophilic substrates. The left part was fully covered with the superhydrophobic coating, and the right part was naked copper surface

图3 超疏水核化点上的气泡脱离. 超疏水点宽度: $100\ \mu\text{m}$, 壁面温度: $T_w=110^\circ\text{C}$

Figure 3 The detachment of bubble from a superhydrophobic pattern. The pattern width is $100\ \mu\text{m}$, and the wall temperature is $T_w=110^\circ\text{C}$

腾过程逐渐变化. 如图4所示, 变化大致可分为3个阶段. 在较低热流密度下, 只有超疏水核化点上起泡(图4(a)). 进一步加大热流密度, 则在裸露铜丝上也会出现起泡. 这些气泡会与超疏水点上的气泡发生相互作用(比如合并), 如图4(b)所示, 但可以看到, 超疏水核化点占有统治地位. 在更高热流密度下, 则可以看到相邻的超疏水核化点之间也会发生大范围的相互作用, 可合并产生跨越疏水点间距的大型汽团(cross-pattern slug)^[11].

为考察换热效率提升情况, 在一个大气压下开展饱和沸腾实验. 图5为带有超疏水点阵的铜丝与光滑铜丝的典型沸腾结果比较. 由两者的沸腾曲线(图5(a))可

图4 超疏水点阵铜丝上的沸腾变化. (a) 离散起泡($T_w=106^\circ\text{C}$); (b) 局部合并($T_w=111^\circ\text{C}$); (c) 跨越点阵合并($T_w=114^\circ\text{C}$)

Figure 4 Different boiling regimes on the superhydrophobic patterned wire. (a) Isolated on-pattern bubbling ($T_w=106^\circ\text{C}$); (b) on-pattern coalescence ($T_w=111^\circ\text{C}$); (c) cross-pattern coalescence ($T_w=114^\circ\text{C}$)

以看出, 带有超疏水点阵铜丝的沸腾起始要早于光滑铜丝, 同时曲线整体较光滑铜丝的沸腾曲线左移, 即其换热效率明显高于光滑铜丝. 图5(b)给出了两者沸腾换热效率的对比. 在热流密度低于 $10^6\ \text{W/m}^2$ 的区间, 超疏水点阵表面的换热效率要比光滑铜丝的高出40%~60%.

可以看到, 超疏水点是优良的核化点, 其有效降低了沸腾起始所需的壁面温度. 本文进一步讨论了超疏水点大小的影响. 如图6所示, 当疏水点大小从20增加到 $1000\ \mu\text{m}$ 时, 起泡所需过热度从 7.3°C 降到 4.2°C , 降低趋势则逐渐趋缓.

图5 (网络版彩色)带有超疏水点阵的铜丝与光滑铜丝的典型沸腾结果比较。(a) 沸腾曲线; (b) 换热效率

Figure 5 (Color online) The comparison of typical boiling results on a superhydrophobic patterned wire and a bare copper wire. (a) Boiling curves; (b) heat transfer coefficients

本文尝试用文献[12~16]中的沸腾换热模型来拟合超疏水点阵铜丝上中低热流密度下的离散气泡核态沸腾换热。从图7可以看出, Forster模型、Rohsenow模型和Stephan模型都较大地低估了沸腾热流, 而Judd模型则稍微高估。与实验数据最为接近的是考虑气泡扰动增强流体换热的Mikic模型。为获得更准确的模型, 进一步基于复合换热模型(partitioning heat transfer model)[17,18]进行分析。该模型在Gerardi等人[19]的实验中表现出色。该模型认为, 总换热由3部分组成, 即气泡扰动造成的流体换热、没有气泡覆盖处表面的自然对流换热, 以及气泡带走的潜热:

$$q''_t = q''_{tc} + q''_{nc} + q''_{la} \quad (1)$$

第一项换热贡献由Mikic模型[15]提供:

$$q''_{tc} = 2\sqrt{\pi f(k\rho c)_l} D_b^2 n \Delta T, \quad (2)$$

其中, f , D_b , n 分别是脱离频率、脱离直径以及气泡点密度; ρ , c 分别为液体密度和比热。

第二项由Churchill公式[20]计算。通过 Nu 数的获得,

图6 起泡所需过热度随超疏水点宽度的变化

Figure 6 Required superheat for bubble nucleation varied with the superhydrophobic pattern width

图7 (网络版彩色)超疏水点阵微细丝上沸腾实验曲线及其与各个模型曲线的对比

Figure 7 (Color online) The experimental boiling curve on the superhydrophobic patterned wire and its comparison with the predictions of various models

可以计算对流换热系数并最终获得式(1)的热流密度:

$$Nu_{nc}^{1/2} = 0.6 + 0.387 \left[\frac{Ra}{1 + (0.599 / Pr)^{9/16}} \right]^{1/6} \quad (3)$$

第三项通过气泡体积计算:

$$q''_{la} = h_{fg} \rho_v V_v f n \quad (4)$$

计算中采用的参数均为已知或实验录像获取的气泡参数。 h_{fg} , ρ_v , V_v , f , n 分别为汽化潜热、蒸汽密度、气泡脱

离体积、脱离频率、起泡密度。如图8所示,该复合模型与实验结果取得很好的一致性。

如前所述,微尺度超疏水点阵表面的换热效率比光滑铜丝的高出40%~60%。结合复合沸腾换热模型,超疏水点阵铜丝换热效率的提高主要有以下两方面原因:首先,超疏水性降低了初始核化所需过热度,使得沸腾在过热度较低的情况下便可发生,因而核化点数目相对于光滑表面增多。在热流密度低于 10^6 W/m^2 的范围内,疏水点阵铜丝上的气泡数目较光滑表面平均增多约22%。其次,相对于光滑表面,超疏水点阵造成的核化在换热面上更加均匀,避免气泡集中生长,实现对整个加热面的充分换热^[21]。而对于每个核化点而言,综合考虑气泡直径和气泡生长频率,并未发现超疏水点比光滑表面上的核化点有明显优势,因此排除其作为换热强化的主要原因。

3 结论及展望

利用影印石版术和非电解电镀沉淀法,在直径为 $150 \mu\text{m}$ 的铜丝上修饰了宽度为 $100 \mu\text{m}$ 、间距为 3 mm 的超疏水点阵。实验证明,超疏水点阵可降低沸腾核化所需过热度。实验观察到了超疏水点阵铜丝上独特的核态沸腾气泡动力学现象,并总结出了热流逐渐增加时的不同沸腾阶段。可以看到,由于更容易起泡,超疏水点在沸腾系统内占据了统治地位,裸露位置的铜丝表面上的起泡基本上都会受到临近超疏水点起泡的影响。

依据复合换热模型,超疏水点阵铜丝上的沸腾换热计算可分为3个部分:自然对流换热、气泡带走潜

图8 (网络版彩色)复合模型^[17,18]与超疏水点阵微细丝上沸腾实验结果的对比

Figure 8 (Color online) Comparison between the heat flux partitioning model^[17,18] and the experimental result of the boiling heat transfer on the patterned wire

热、气泡扰动换热。其中,扰动换热是总换热的主要部分。经超疏水点阵修饰的铜丝的核态沸腾换热效率较光滑铜丝提高了40%~60%。相较于光滑铜丝,超疏水点阵铜丝更多的核化点及更均匀的空间分布提高了气泡换热效率。但是,核化点的最佳密度以及排布还有待澄清。固体壁面的特征尺寸、导热及热容与该最佳密度的设计应该有密切联系,有待进一步研究。

参考文献

- Wang C H, Dhir V K. Effect of surface wettability on active nucleation site density during pool boiling of water on a vertical surface. *J Heat Transf-Trans ASME*, 1993, 115: 659-669
- Takata Y, Hidaka S, Cao J M, et al. Effect of surface wettability on boiling and evaporation. *Energy*, 2005, 30: 209-220
- Nam Y, Ju Y S. Bubble nucleation on hydrophobic islands provides evidence to anomalously high contact angles of nanobubbles. *Appl Phys Lett*, 2008, 93: 103115
- Phan H T, Caney N, Marty P, et al. Surface wettability control by nanocoating: The effects on pool boiling heat transfer and nucleation mechanism. *Int J Heat Mass Transf*, 2009, 52: 5459-5471
- Betz A R, Xu J, Qiu H, et al. Do surfaces with mixed hydrophilic and hydrophobic areas enhance pool boiling? *Appl Phys Lett*, 2010, 97: 141909
- Betz A R, Jenkins J R, Kim C J, et al. Significant boiling enhancement with surfaces combining superhydrophilic and superhydrophobic patterns. In: International Conference of Micro Electronics and Mechanical Systems (MEMS). Cancun, 2011
- Jo H J, Ahn H S, Kang S H, et al. A study of nucleate boiling heat transfer on hydrophilic, hydrophobic and heterogeneous wetting surfaces. *Int J Heat Mass Transf*, 2011, 54: 5643-5652
- Wang X, Zhao S, Wang H, et al. Bubble formation on superhydrophobic-micropatterned copper surfaces. *Appl Therm Eng*, 2012, 35: 112-119
- Betz A R, Jenkins J, Kim C J, et al. Boiling heat transfer on superhydrophilic, superhydrophobic, and superbiphilic surfaces. *Int J Heat Mass*

- [Transf](#), 2013, 57: 733–741
- 10 Larmour I A, Bell S E J, Saunders G C. Remarkably simple fabrication of superhydrophobic surfaces using electroless galvanic deposition. *Angew Chem Int Edit*, 2007, 46: 1710–1712
- 11 Wang X W, Song Y X, Wang H. Observation of nucleate boiling on a fine copper wire with superhydrophobic micropatterns. *Chin Phys Lett*, 2012, 29: 114702
- 12 Rohsenow W M. A method of correlating heat transfer data for surface boiling of liquids. *J Heat Transf-Trans ASME*, 1952, 4: 969–975
- 13 Forster K E, Greif R. Heat transfer to a boiling liquid—Mechanism and correlations. *J Heat Transf-Trans ASME*, 1959, 81: 43–52
- 14 Stephan P, Hammer J. A new model for nucleate boiling heat transfer. *Heat Mass Transf*, 1994, 30: 119–125
- 15 Mikic B B, Rohsenow W M. A new correlation of pool-boiling data including the effect of heating surface characteristics. *J Heat Transf-Trans ASME*, 1969, 91: 245–250
- 16 Judd R L, Hwang K S. A comprehensive model for nucleate pool boiling heat transfer including microlayer evaporation. *J Heat Transf-Trans ASME*, 1976, 98: 623–629
- 17 Kurul N, Podowski M Z. Multidimensional effects in forced convection subcooled boiling. In: *Proceedings of 9th International Heat Transfer Conference*. Jerusalem, 1990
- 18 Bowring R. Physical Model, Based on Bubble Detachment, and Calculation of Steam Voidage in the Subcooled Region of a Heated Channel. Report No. HPR-10, Institutt for Atomenergi, Norway, 1962
- 19 Gerardi C, Buongiorno J, Hu L, et al. Study of bubble growth in water pool boiling through synchronized, infrared thermometry and high-speed video. *Int J Heat Mass Transf*, 2010, 53: 4185–4192
- 20 Churchill S W, Chu H H S. Correlating equations for laminar and turbulent free convection from a horizontal cylinder. *Int J Heat Mass Transf*, 1975, 18: 1049–1053
- 21 Han C Y, Griffith P. The mechanism of heat transfer in nucleate pool boiling—Part I. *Int J Heat Mass Transf*, 1965, 8: 887–904

Summary for “带有超疏水点阵的微细铜丝上的核态沸腾”

Nucleate boiling on thin copper wires with micropatterned superhydrophobic and hydrophilic areas

Xinwei Wang & Hao Wang*

College of Engineering, Peking University, Beijing 100871, China

* Corresponding author, E-mail: hwang@coe.pku.edu.cn

Boiling is widely employed in practical applications due to its excellent heat-transfer characteristics. To meet the ever-increasing demand of heat dissipation, boiling optimization is an important topic in heat transfer research.

In this work, saturated pool boiling experiments were conducted on thin copper wires with superhydrophobic micropatterns. The investigation was conducted on a thin copper wire patterned with superhydrophobic sites. The wire was 150 μm in diameter and 85 mm in length. Briefly, the wire was first uniformly coated with a photoresist. The patterns were then created by a means of soft lithography using with a photo-plotted mask. Afterwards, an electroless galvanic deposition method was applied to coat a thin superhydrophobic layer onto the exposed copper patterns, by consecutively immersing the wire in AgNO_3 and heptadecafluoro-1-decanethiol (HDFT) solutions. Finally, the remaining photoresist on the wire was removed, leaving the superhydrophobic micropatterns coated onto the copper surface. In most tests, the patterns were 100 μm wide with 3-mm separation distance. De-ionized (DI) water was used as the working fluid, and boiled and degassed for 30 minutes in the chamber prior to the measurement. The testing wire was also heated in the chamber to degas. A high-speed CCD camera up to 30000 frames per second was mounted on a microscope for continuous imaging of bubble formation. Optical illumination was achieved by a fiber optic illuminator system.

The superhydrophobic pattern lowered the superheat for bubble nucleation. Larger pattern area led to lower superheat. The superheat dropped from 7.3 to 4.2°C when the pattern width increased from 20 to 1000 μm .

When bubble grew on the pattern site, there was a neck region connecting the detaching bubble and the pattern. As the bubble detached with the breakage in the middle of the neck region, a residual bubble still remained, which served as the embryo for the next cycle. However, the existence of the embryo didn't significantly facilitate the bubbling process itself, as compared to that over the bare copper region.

The bubble interactions on the superhydrophobic micropatterns were studied. Different boiling regimes were observed as the heat flux increased: (1) Isolated on-pattern bubbling regime; (2) on-pattern coalescence regime; (3) cross-pattern coalescence. The superhydrophobic micropatterns played dominant roles in bubble formation as well as the bubble interactions. The bubbles on the bare copper surface could not grow independently.

Heat transfer efficiency was enhanced by 40%–60% as compared to that on the bare copper wire. The wettability patterning on the wire significantly lowered the nucleation temperature, while increasing the bubble density. However, no appreciable change over the bubble detachment volume and frequency on single nucleation site was observed. Based on the observed bubble parameters, the nucleate boiling heat transfer can be predicted and calculated by using the heat flux partitioning model. The heat transfer comprised of three components: Natural convection, latent heat of bubble growth, and transient conduction caused by bubble growth and detachment. The third mechanism was found to be the major contributor to the heat transfer in the studied nucleate boiling on the patterned wires.

Future work is needed to clarify the optimal distribution of the patterns, which is supposed to be related to the wire geometry, conductivity and capacity.

nucleate boiling, hydrophilic, superhydrophobic, bubble, heat transfer enhancement

doi: [10.1360/TB-2019-0518](https://doi.org/10.1360/TB-2019-0518)