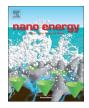
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# Nano Energy



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# Enhanced bubble nucleation and liquid rewetting for highly efficient boiling heat transfer on two-level hierarchical surfaces with patterned copper nanowire arrays

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## ARTICLE INFO

Keywords: Boiling heat transfer Two-level hierarchical surface Copper nanowires Capillary pumping Bubble nucleation Liquid rewetting

## ABSTRACT

Enhancing boiling heat transfer by surface modification is of critical interest for improving the efficiency of many energy systems and for addressing thermal management bottlenecks in electronics. However, the improvement of all boiling heat transfer characteristics including the critical heat flux, heat transfer coefficient and onset of nucleate boiling, usually has conflicting requirements on surface wettability and morphology. In this work, we develop a two-level hierarchical surface with patterned copper nanowire arrays for boiling heat transfer enhancement. By surrounding long nanowire arrays with short nanowires where microcavities are formed between short nanowire clusters, a novel strategy is reported to improve all the boiling heat transfer characteristics through increasing bubble nucleation site density, capillary-induced liquid rewetting, and the separation of liquid-vapor pathways. Compared to boiling heat transfer performance on the plain copper surface, a 71% higher critical heat flux, a 185% higher heat transfer coefficient as well as a 37% lower onset of nucleate boiling are demonstrated on such two-level hierarchical surfaces. In addition, we correctly predict the effect of surface structure on the boiling heat transfer performance by an analytical model. Through distinguishing the role of different structure morphologies including the improved nucleation site by microcavities, enhanced liquid wicking by nanowires, and continuous liquid supply by long nanowire arrays, we have established a comprehensive understanding on the relation between the surface structures and boiling heat transfer characteristics.

### 1. Introduction

Boiling heat transfer is of great importance for a broad range of industrial applications ranging from thermal power plants and air conditioners to thermal management of electronics [1-6]. The efficiency of boiling process is quantified by the heat transfer coefficient (HTC), defined as the ratio of the heat flux to wall superheat which measures the temperature difference between the solid surface and bulk liquid. The HTC usually increases with increasing heat flux; but beyond a critical heat flux (CHF) a vapor film develops between the solid surface and bulk liquid, which severely limits heat transfer performance, a phenomenon referred as the boiling crisis [7–10].

The ultimate goal for enhancing boiling heat transfer is to maximize CHF and increase HTC simultaneously. In addition, it is necessary to reduce thermal loads required for the onset of nucleate boiling (ONB), which represents the beginning point of more efficient nucleate boiling compared to single-phase convective heat transfer.

Many efforts have been devoted to improving boiling heat transfer by modifying the properties of working fluids [11–13] and boiling surfaces [14–16], and by introducing active techniques such as applying electric fields and ultrasonic agitation [1,17,18]. Surface modifications are particularly attractive because often there are constraints on the implementation of working fluids and the operational conditions to introduce active techniques. Surface wettability and

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http://dx.doi.org/10.1016/j.nanoen.2017.05.028 Received 17 March 2017; Received in revised form 8 May 2017; Accepted 11 May 2017 Available online 22 May 2017 2211-2855/ © 2017 Elsevier Ltd. All rights reserved.



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morphology, which are inter-related, are the two major factors determining the boiling heat transfer performance of the structured surfaces [19,20]. To improve HTC, surface wettability is typically reduced to promote vapor generation through improved bubble nucleation due to the lower energy barrier for nucleation on a surface with poor wettability (large contact angle for bulk liquid) [21,22]. However, too many nucleation sites can be detrimental at high heat flux, where densely packed bubbles coalesce and form a vapor film limiting the further increase of CHF. As such, many boiling surfaces were designed to suppress bubble nucleation by merely increasing wettability, i.e., reducing contact angle, and thus increasing CHF. Consequently, the increased wettability (small contact angle) lowers HTC and increases ONB in low heat flux range due to more energy being dissipated by convection rather than vapor generation. Apparently improving all boiling heat transfer characteristics (CHF, HTC, and ONB) has distinctly conflicting requirements on the surface wettability; and it has, to date, remained challenging to develop a robust surface that can improve all the key boiling characteristics.

With the advancement of micro/nano-fabrication, numerous structured surfaces have been explored to improve boiling heat transfer performance [23-25]. Since the bubble nucleation size is in the microscale, various microstructures such as microcavities [26], microchannels [27,28], micropillars [29,30], micromeshes [31,32], and microporous surfaces [33,34] have often been used to modify surface morphology to increase the bubble nucleation site density for higher HTC. Additionally, notable increases in HTC have also been reported using embossed microstructures with specific contoured shapes, which enables evaporation momentum to promote the separation of liquid and vapor flow paths and thus enhancing micro-convection [35,36]. Despite that nanoscale structures are not expected to improve bubble nucleation sites, such surfaces have been exploited to enhance boiling heat transfer by promoting liquid rewetting and heat transfer area [37–39]. The separations between nanostructures provide a very large capillary force, such structures can act as efficient wicking surfaces for liquid rewetting. In addition, the effective heat transfer area of nanostructured surfaces is dramatically higher than that of microstructured surfaces due to the larger specific surface area. More importantly, the microscale defects formed due to the clustering of nanostructures during the drying process of surface preparation can serve as potential bubble nucleation sites [6,40-42]. However, due to the merging of the dense bubbles at high heat flux, a vapor film forms between the solid surface and bulk liquid and it expands arbitrarily on the entire boiling surface, which interrupts liquid rewetting and hinders further improvement of CHF. Clearly, the boiling heat transfer characteristics are closely related to the bubble behaviors including bubble nucleation, growth, coalescence, and departure.

Inspired by the demonstration of enhanced bubble nucleation by microcavities [42–44] and improved liquid rewetting by nanostructures [3,6,16,45], as well as the recent efforts in separating liquid-vapor pathways [36,46,47], in this work we develop a two-level hierarchical surface composed of long nanowire arrays (the first level) and short nanowire clusters (the second level) to improve boiling heat transfer. By manipulating bubble dynamics with different structure morphologies, microcavities (1–10  $\mu$ m) for efficient bubble nucleation and microvalleys (100  $\mu$ m) for the separation of liquid-vapor pathways, the hierarchical surface can improve all the boiling heat transfer characteristics. Compared to boiling heat transfer on the plain copper surface, a 71% higher CHF, a 185% higher HTC as well as a 37% lower ONB are demonstrated on such two-level hierarchical surface.

#### 2. Experimental section

#### 2.1. Preparation of boiling surfaces

Due to the wide applications of copper in thermal systems, high purity copper (99.9% purity) was used to fabricate the test samples in

this work. Each test sample was polished by 3000 grit sandpaper, cleaned in an ultrasonic bath with acetone for 10 min, and then rinsed with isopropyl alcohol, ethanol, and deionized (DI) water. The samples were then dipped into a 2.0 M hydrochloric acid solution for 10 min to remove the native oxide film on the surface, then triple-rinsed with DI water, and dried with clean nitrogen gas. For comparison, we fabricated a plain copper surface and three nanowired copper surfaces including a two-level hierarchical surface and two surfaces with uniform nanowires in the length of 5 µm and 30 µm, respectively. Uniform copper nanowires on the surface were fabricated by a two-step porous anodic alumina oxide (AAO) template-assisted electro-deposition method [48]. The porous AAO template was first bonded onto the sample surface by electrodepositing at -0.8 V for 15 min. During the deposition process, short nanorods (~ 1 µm) were formed to serve as the screws to connect the porous AAO template and the sample. In the second step, copper nanowires were grown by depositing copper in a three-electrode electroplating cell with the same electrolyte as that in the first step (Supplementary Section 1). The length of copper nanowires was controlled by the electroplating time. Here, the nanowires with an average height (length) of 5 µm and 30 µm were grown by the electroplating for 30 min and 240 min, respectively. After immersed in 1 M NaOH solution to dissolve the porous AAO templates and cleaned with DI water, uniform nanowired surfaces were obtained.

Fig. 1 shows the fabrication process of a two-level hierarchical surface with patterned copper nanowires in different lengths. A standard photolithography and wet-etching method was chosen to first fabricate the copper micropillars, as shown in Fig. 1a. Copper nanowires were then grown on the micropillared surface using a twostep porous AAO template-assisted electroplating method, as shown in Fig. 1b. Due to the different growth time for the nanowires on the micropillared surface, long nanowire arrays were grown on top of micropillars while short nanowires were formed in the microvalleys between micropillars (Supplementary Section S1).

#### 2.2. Surface characterization

The field-emission scanning electron microscopy (FE-SEM, JEOL JSM-7401F) was employed to examine the morphologies of the plain copper surface and nanowired copper surfaces. The contact angle of a 5  $\mu$ L water droplet on the surface was measured using an optical imaging system. The contact angles on each test surface were measured and averaged over six measurements.

#### 2.3. Boiling heat transfer experiments

Boiling heat transfer experiments were conducted using a custombuilt boiling heat transfer testing system (Supplementary Section 2). The system consisted of a boiling chamber, a heating system, and a data acquisition system. The chamber had a diameter of 19 cm and a height of 22 cm and was directly open to the ambient atmosphere. An electrical resistance heater served as the heat source to maintain the saturation condition of DI water in the chamber. Each test sample with a thickness of 0.8 mm was diced to a projected surface area of 8  $\times$ 8 mm<sup>2</sup> and then it was soldered onto the copper heating bar (Fig. S5). Three holes were drilled into the center of the heating bar at 8 mm intervals. Three K-type thermocouples (± 0.2 K) were installed to measure the temperature distribution in the heating bar. The steady state measurement method was used to measure the boiling heat transfer performance. All tests were performed using degassed DI water to avoid premature bubble formation and to minimize surface contamination. The input power started from 1 W cm<sup>-2</sup> and was increased with the increment of 20 W cm<sup>-2</sup> in the initial stage and 5 W cm<sup>-2</sup> when it was close to CHF. All values of the temperature and pressure were collected by the data acquisition to determine the uncertainty of heat flux, superheat, and heat transfer coefficient (Supplementary Section 3).

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