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EXPERIMENTAL STUDY OF NUCLEATE BOILING PERFORMANCE ON SEVERAL TYPES OF THE MIXED-WETTABILITY PATTERN SURFACE BY MICRO/MILLI-SIZED HYDROPHOBIC PATTERNS

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ABSTRACT

To increase the nucleate boiling efficiency, many nucleate boiling experiments have been conducted and could get brilliant and challengeable results. A consensus was that CHF and heat transfer were affected by a modified heating surface which change the micro roughness, thermophysical properties of heating surface, or the wettability. Of the many parameters, the wettability study is regarded as the most powerful factor. For finding the optimized condition at the nucleate boiling (high heat transfer and high CHF), we design the special heaters to examine how two materials, which have different wettabilities, affect the boiling phenomena. The special heaters have several types of hydrophobic patterns which have the precise size because they were made by MEMS techniques on the silicon oxide surface. In the experiments with patterned surface, hydrophobic dots lead to an early bubble inception and induce the better heat transfer. These experiments are compared with classic and recent models for bubble inception. The all experiments are conducted under the saturated pool boiling condition with distilled water at 1 atm pressure. The peculiar Teflon (AF1600) is used as the hydrophobic material. The hydrophilic part is performed by silicon oxide through the furnace procedure. The experiments using the micro-sized patterns and milli-sized patterns are performed, and the results are compared with the reference surface. These mixedwettability studies are expected to induce the development of the nucleate boiling condition.

INTRODUCTION

The problem of cooling has become increasingly critical in the nuclear industry. The most effective way of cooling a Soon Ho Kang POSTECH Pohang city, Republic of Korea

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nuclear power plant running at high temperatures is boiling heat transfer, which exploits the latent heat of vaporization during the phase change from liquid to gas. This boiling mode is called nucleate boiling regime. At the nucleate boiling regime, two parameters are regarded as most important factors; Critical Heat Flux (CHF), Heat Transfer Coefficient (HTC). The CHF is a serious limitation in various apparatuses that use boiling phenomena. If an operating condition is over the CHF point, the operating equipment will have a low efficiency or be even failed under a worst condition. On the other hand, the enhancement of nucleate boiling heat transfer coefficient enables the equipment to be operated in a higher performance under the same limitation.

Based on this motivation, many pool boiling experiments have been conducted for the enhancement of the boiling condition over the past several years. It has produced brilliant and challengeable results that the boiling condition (such as CHF and nucleate boiling heat transfer) is governed by the condition of the heating surface. One of them is the nanofluds experiment which exhibits an incredible enhancement of the CHF when used as a working fluid in pool boiling. It has been proved that the outstanding the CHF enhancement is due to the changed surface [1, 2]. In this consideration, Kim et al. [3] concluded that the CHF is enhanced by the wettability, surface geometry, and capillary wicking effect. It well correspond to prior researches [4, 5, 6]

The enhancement of nucleate boiling heat transfer is also considered as a hot issue because it enables the equipment to be operated in a higher performance under the same limitation. So many pool boiling experiments have been conducted for the

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enhancement of the boiling condition over the past several years. Based on these previous studies, the impacts of heating surface characteristic are treated as very important. In this consideration, many researches were contributed to improve boiling condition with various methods such as coating the porous media [7], making the surface micro-structure [8, 9] or changing the roughness [10, 11]. Among those methods, the change of wettability is regarded as a powerful method. Through an oxidation method, researchers have shown that the lower contact angle of the heating surface, the less nucleate boiling density.[12] The Yang and Kim[13] also supported the trend of wettability effect by their mathematical model of the pool boiling nucleation site density. Hibiki and Ishii [14] also showed that the nucleation site density is dominantly related with the wettability of heating surface. Recently, Phan et al. [15] also conducted pool boiling experiment on various wetting surfaces. In this respect, the excellent boiling performance (high CHF and high nucleate boiling heat transfer) in pool boiling could be achieved by the modified surface such a favour way that it satisfies the optimized wettability condition. In a similar approach, Takata et al experimentally studied the pool boiling with checked and spotted patterns surface made of a super hydrophobic material [16]. Their brilliant and amazing challenge brought a fairly new idea for understanding the wettability effect on boiling phenomena, but the experiment involved two limitations. One was that the micro-scaled height difference between the super hydrophobic material and bare surface affected the boiling phenomena. Another was that the experiments were conducted in a milli-dot order (from 3mm to 5mm). Most recently, Nam and Ju [17] developed the special surface which consisted of hydrophobic dot and hydrophilic basic surface without any micro geometry. Even though Nam and Ju reported interesting result that earlier bubble inception is induced by hydrophobic dot, the experiment couldn't cover the total nucleate boiling heat transfer performance.

So we conducted the heat transfer pool boiling experiments on hydrophobic dots, fabricated on hydrophilic surface, to study the effect of wettability about total nucleate boiling heat transfer on the several kinds of hydrophobic pattern arrangements without micro structure.

NOMENCLATURE

q" heat flux (w/m²). T_{wall} Wall supherheat (K)

EXPERIMENTS

Experiment Apparatus

The test apparatus is designed for a pool boiling experiment under atmospheric pressure by electronic joule heating method. It consists of a test sample jig, a main test pool, and a lid with an immersion heater and a condenser.



Figure 1 Schematic diagram of pool boiling facility (a) reflux condenser (b) immersion heater (c) Heating sample part and (d) Constant temperature vessel and reference resistance.

The test sample jig is designed to hold the test sample in the test pool and to make changing the test sample. The jig consists of and anodized aluminium cylinder base and a PEEK (polyetheretherketone) test sample frame. PEEK is a thermoplastic which has high thermal resistance and is compatible with an aqueous environment. The base and the frame are connected by bolts and O-rings. The test sample frame has a framework for sample mounting. Test samples were fixed and waterproofed with adhesive sealant (Permatex, clear RTV silicone) and binary epoxy (DuralcoTM, 4461-IP).

The main test pool is an octagonal aluminium cylinder bath with four glass windows. It is 165mm high and has a 3 liter capacity. All side walls except for the glass windows of test pool are thermally insulated with a heating insulating material (AEROFLEX, Eastern Polymer Industry Company). The test sample jig can be interconnected and separated before and after each test, through a large hole on the bottom of pool. The main test pool is connected to the aluminium lid plate by bolts and O-rings. The lid is an aluminium plate which supports an immersion heater below it and a connector for a reflux condenser above it. The immersion heater is operated by a PID (Proportional Integral-Derivative) controller to maintain the pool at the saturation temperature by detecting the bulk temperature with a K-type thermocouple. A reflux condenser prevents water loss from the test pool by evaporation, and maintains atmospheric pressure inside the pool. Tap water was used as the reflux condenser coolant.

Experiment Procedure

All experiments were conducted on saturated condition with di-stilled water. By using PID temperature controller, the temperature of the bulk water in the boiling pool was maintained at saturated temperature (100°C). Before starting the main experiment, the used working fluid experienced preboiling procedure to degas during 3 hours. With the system still open to the ambient through the reflux condenser,, the escaping air is pushed away while the steam is condensed back to the vessel. The test was conducted by increasing the electric power



Figure 3 Details of the test sample design.

supplied to the test sample in small steps. Each step was gained from 200 data points in a quasi-steady state.

Heating surface preparation

To facilitate both the heating and surface modification, a thin film heater was embedded on one side of a silicon wafer and artificial surfaces were created on the other side of the wafer using the microelectromechanical systems (MEMs) technique. The test heater is a rectangular silicon wafer plate. The substrate silicon plate is 25mm x 20mm, and has a SiO₂ layer to eliminate the native oxidation effect on both the top and bottom. The heating part was a platinum thin film of about 1500 Å thickness layered on the bottom of the substrate using an E-beam evaporator. The titanium thin film is used as the intermediate layer between the silicon wafer and platinum. The titanium played as role of adhesion layer between SiO₂ and Pt. To prevent the effect of unpredictable side bubbles, which is not a result of heating surface condition, on boundary of sample mount, the real heating area (Pt) is smaller than the size of sample (Si). The complete platinum film heater has an H shape because they consist of electrode part and main heating part. The centre region of the H shape is 15mm x 10mm which is main heating area. The electrode part is filled with lead by soldering to attach wire connection. With lead soldering, their resistance of electrode part becomes much smaller than 1% as compared with total sample resistance. It is natural that the heating area is defines as 15mm x 10mmm. The heat loss calculation was conducted by computational analysis.

The modified surfaces consist of two different wettability characteristics; the hydrophobic coating part and hydrophilic substrate part. The hydrophilic substrate part is the SiO₂ which has the 5000 Å thickness by oxidation procedure in furnace. The particular Teflon AF1600 (Dupon polymer, Inc) is used as the hydrophobic coating material. The total fabrication

procedure is based on the Lift-off method of MEMs technique to fabricate precise micron-sized pattern shape. The AF1600 is dissolved into a FC40 (3M Inc.) solvent to advance the MEMs patterning. With this especial solvent, the total patterning fabrication is progressed. First of all, the photoresist (PR) is patterned with designed mask on a silicon substrate which is already oxidized through furnace. And then the special solution, which is mixed the AF1600 with the FC40, is pin coated on the developed samples. After spin coated process, test sample suffer evaporating procedure to eliminate the FC40. Lastly by using acetone and methanol, the photoresist and the part of Teflon, which is coated on undersigned, is removed on silicon wafer. In this paper, four kind of pattern sample is used as heating surface. The diameters of 100µm and 1mm are fabricated and each hydrophobic island is separated from each other by more than three times of each pattern diameter to prevent any interactions among bubble nucleated. (Zhang and Shoji [18]) Table 2 contains the information about heating condition and arrangement of hydrophobic dots.

Property		Bare	Teflon	Pattern		
Contact Angle (°)		54	123	Each value is preserved		
Roughness	Ra	1.75	1.55	4.38		
(nm)	Rt	17	27.04	52.76		
Table 1 Information of surface characteristic.						



Figure 4 The contact angle (a) on SiO2, (b) on Teflon, (c) 3D profiler image about Teflon pattern (d) on Teflon dot

Туре	Micro Dot		Milli Dot	
Expression	A1	A2	B1	B2
Diameter	100µm	100µm	1mm	1mm
Area ratio = Teflon total area/ heating area	3.14%	1.57%	3.14%	1.57%
Number of dot	600	150	6	3
Pitch	400µm	400µm	3mm	3mm

Table 2 Information of hydrophobic patter surface.

Surface Characterization

We investigate the static contact angle to know the exact difference on the surface wettability. 3μ l water droplet is used to generate a droplet meniscus on each surface. The measuring is repeated 3 times to guarantee the surface reproducibility on each condition. The result of repeated measurements indicates that the measuring uncertainty is about 3°. The contact angle of the silicon dioxide surface, which contains 5000 Å, is 54°. The hydrophobic surface, made by the Teflon, has 123°. Also it is confirmed that the surface pattern procedure couldn't change the static contact angle by comparing the measured value. All values of contact angle were measured at room temperature (20°C).

The roughness of heating surface is measured by 3D profiler. Among many roughness parameters, we focus on the Ra (arithmetical average roughness) and parameter Rt. The Ra is one of the roughness parameter most commonly used so far and the Rt can suggest the maximum difference height on heating surface. It is confirmed by the 3D profiler that bare surfaces and Teflon patter surfaces does not possess a micro structure which can entrap the nucleus of bubble initiation. The average Ra and Rt of several points are 1.75nm and 17nm on bare surface. The roughness on hydrophobic whole coating surface has almost similar value. The roughness of Teflon pattern has larger than bare surface but not to so much as to entrap the vapour nucleus. The Ra and Rt of Telfon pattern are 4.38nm and 52.76nm. Table 1 includes the detailed information about wettability and roughness.

Experimental Result and Discussion

Before the start main experiment, we conducted the reproducibility experiment for reliability of our experimental facility. All experiments are conducted on plate pool boiling under 1 atm saturated condition with degassed di-water. Consequently, laid out as shown in figure 5, we can get the well repeated data. It is sure that the reproducibility is enough to analyze the certain trend.

A notable point of the boiling experiment on bare surface is that the heterogeneous boiling is generated on the nano-scale roughness surface which is assured at previous section. By observation through visualization window, the ONB (Onset of Nucleate Boiling) is occurred when the surface temperature is near 112°C. It is contrary to the expectations of classic bubble nucleation. As previous studies, the cavity roughness played as an important role on bubble nucleation. However the roughness of present heating surface, used in this paper not over than 20nm on bare surface, is too low to act as a bubble nucleation site. According to the prediction of Blander and Katz [19], who discussed about the bubble nucleation on very smooth surface by including hydrodynamic and diffusion constraints, the predicted ONB on current surface is over 300K. By the Clausius-Clapeyron relation and Young-Laplace equation, the ONB of present surface is over 1000K on bare surface. Even though these condition, the similar phenomena on nanometerscale roughness is observed by Theofanous et al[20] with diwater on highly controlled Ti surface and Qi and Klausner[21] with ethanol on ultra smooth metallic surface. Based on these observations and our experiment results, we conclude that the nano-scale roughness is enough condition to initiate the heterogeneous bubble nucleation.



Figure 5 Experimental reproducibility.



Fig ure 6 experimental boiling curves of (a) $100\mu m$ pattern and (b) 1mm pattern.

Now attention is focused on the boiling curve on several pattern conditions. Figure 6 shows the result of boiling curve on several pattern conditions. As mentioned in previous section, the symbol 'A' and 'B' mean that the diameter of hydrophobic dot is 100µm and 1mm. And the number '1' and '2' indicate that the ratio between the total Teflon area and heating surface area is 3.14% and 1.57%. So a number of hydrophobic dots on one heating surface is controlled to restrain prior conditions. For focusing on nucleate boiling heat transfer performance, the CHF of each sample isn't contained in figure 6.

According to boiling curves, the hydrophobic pattern induces the better heat transfer performance than the bare surface. Some researchers tried to depict the early bubble inception on hydrophobic surface. They got results which can explain some phenomenon. But it has the limitation. First, the previous studies started from the existence of cavitations on heating surface. To act a bubble nucleation site, the size of cavities should be greater than the critical radius, which is in the order of the micrometer for water. But in this paper, the micron-cavities, the place of bubble initiation by previous theories, do not exist. Second, they couldn't quantitatively predict exact bubble initiation temperature. So in this paper, we tried to different approach. It is conjectured that the reason of enhancement results from the characteristic of hydrophobic surface. By several physicists, the existence of nanobubbles, have a radius of curvature of the order of several hundreds nm, and a height above the substrate of several tens nm, is reported. [22-26] Nam and Ju [17] also reported the incipience of nanobubbles at low superheat on micron size hydrophobic patterns. So it is conjectured that the nanobubbles on hydrophobic pattern enhance the nucleate boiling heat transfer ability by activating at lower superheat than on bare surface which couldn't contain the nanobubbles.

Of the hydrophobic pattern surfaces, the highest rate of heat transfer occurs on 100µm diameter. Among them, the higher area ratio condition is the best. And contrary to the prior expectation, 1mm hydrophobic dots couldn't enhance the boiling heat transfer performance. The cause of this difference between the pattern surfaces is expected to come from the difference of a number of hydrophobic dots. Based on the nanobubbles theories, the hydrophobic patterns have the possibility to be activated nucleation site even though they don't have any micro structures. In the case of 'B', a number of hydrophobic patterns is not much than bare surface. So it is conjectured that the enhancement of total nucleate boiling heat transfer is insignificant by just 6 or 3 hydrophobic dots. On the other hand, the case of 'A' has much larger number of hydrophobic dots than the case of 'B'. Consequently it is supposed that the larger number of hydrophobic dots had a chance to enhance the activated nucleation site density which is directly related with the total nucleate boiling heat transfer performance.

CONCLUSION

The nucleate boiling experiment on highly controlled surfaces with the special surface characteristics is conducted, and we conclude that

- The highly controlled surface by MEMs technique has good reproducibility in our plate pool boiling experiment facility. Notable is that the earlier ONB is occurred than the prediction of classic vapor trapped theory. On the basis of our experiments, finally we conclude that the nano-scale roughness is enough condition to initiate the heterogeneous boiling phenomena.
- The nucleate boiling heat transfer on hydrophobic surface is confirmed higher than that on the bare surface. The reason of increase is conjectured that the nanobubbles on hydrophobic patterns are played as a role of bubble nucleus.
- By comparing the boiling heat transfer performance of case 'A' and case 'B', we reach the conclusion that the sufficient hydrophobic patterns is needed to enhance the nucleate boiling heat transfer ability. Finally it is conjectured that if a number of the hydrophobic dot isn't enough, the nucleate boiling heat transfer will not show the enhancement even as the surface has the hydrophobic dots.

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REFERENCES

- [1] Kim, H.D., Kim, J. and Kim, M.H., 2006, Effect of nanoparticles on CHF in pool boiling of nano-fluids, *Int.J of Heat and Mass Transfer*, 49, pp. 5070-5074.
- [2] Kim, H.D. and Kim, M.H., 2007, Effect of nanoparticle deposition on capillary wicking that influences the critical heat flux in nanofluids, *Appl. Phys. Lett.*, 91, 014104.
- [3] Kim, S.T., Kim, H.D., Kim, H., Ahn, H.S., Jo, H.J., Kim, J. and Kim, M.H., 2009, Effects of nano-fluid and surfaces with nano structure on the increase of CHF, *Experimental thermal and fluid science*, Available online.
- [4] Hahne, E. and Diesselhorst, D., 1978, Hydrodynamic and surface effects on the peak heat flux in pool boiling, *Proceedings of the 6th International Heat Transfer Conference*, Vol. 1, pp. 209-214.
- [5] Liaw, S.P. and Dhir, V.K., 1986, Effect of surface wettability on transition boiling heat transfer from a vertical surface, *Proceedings* of the 8th International Heat Transfer Conference, Vol. 4, pp. 2031-2036
- [6] Golobic, I. and Ferjancic, K., 2000, The role of enhanced coated surface in pool boiling CHF in FC-72, *International Journal of Heat and Mass Transfer*, vol. 369, pp. 525-531

- [7] Liter, S.G. and Kaviany, M., 2001, Pool-boiling CHF enhancement by modulated porous-layer coating: theory and experiment, *Int. J Heat Mass Transfer*, 44, pp. 4287–4311.
- [8] Anderson, T. M. and Mudawar, I., 1989, Microelectronic cooling by enhanced pool boiling of a dielectric fluorocarbon liquid, *Journal* of *Heat Transfer*, vol. 111, pp. 752-759.
- [9] Messina, A. D. and Park, E. L. Jr., 1981, Effects of precise arrays of pits on nucleate boiling, *International Journal of Heat and Mass Transfer*, Vol. 24, pp. 141-145.
- [10] Ferjancic, K. and Golobic, I., 2002, Surface effects on pool boiling CHF, *Experimental Thermal and Fluid Science*, Vol. 25, pp. 565-571.
- [11] Fong R.W.L., McRae G.A. and Coleman C.E., 2001, Correlation between the critical heat flux and the fractal surface roughness of zirconium alloy tubes, *Enhanced Heat Transfer*, Vol. 8, pp. 137-146
- [12] Wang C. H. and Dhir V. K., 1993, Effect of surface wettability on active nucleation site density during pool boiling of water on a vertical surface, *J. Heat Transfer*, Vol. 115, pp. 659–669.
- [13] Yang S. R. and Kim R. H., 1988, A mathematical model of the pool boiling nucleation site density in terms of the surface charateristics, *Int. J Heat Mass Transfer*, 31, pp. 1127-1135
- [14] Hibiki T. and Ishii M., 2003, Active nucleation site density in boiling systems, *Int. J Heat Mass Transfer*, 46, pp. 2587-2601
- [15] Phan H. T., Caney N., Marty P., Colasson S., and Gavillet J., Surface wettability controlled by nanocoating: The effects on pool boiling heat transfer and nucleation mechanism, *Int. J Heat Mass Transfer*, vol. 52, pp. 5459-5471
- [16] Takata, Y., Hidaka, S., Masuda, M. and Ito, T., 2003, Pool boiling on a superhydrophilic surface, *International Journal of Energy Research*, vol. 27, pp. 111-119.
- [17] Nam Y. and Ju Y. S., 2008, Bubble nucleation on hydrophobic islands provides evidence to anomalously high contact angles of nanobubbles, *Appl. Phys. Lett.*, vol. 93, 103115.
- [18] Zhang L. and Shoji M., Nucleation site interaction in pool boiling artificial surface, *Int. J Heat Mass Transfer*, vol. 26, pp. 513-522
- [19] Blander M. and Katz J. L., Bubble nucleation in liquids, *AlChE Journal*, vol. 21, pp. 833-848.
- [20] Theofanous, T.G., Tu, J.P. and Dinh, T.N., 2002, The boiling crisis phenomenon Part I: nucleation and nucleate boiling heat transfer, *Experimental thermal and fluid science*, vol.26, pp. 775-792.
- [21] Qi, Y. and Klausner, J.F., 2006, Comparison of nucleation site density for pool boiling and gas nucleation, *J. Heat Transfer*, vol. 128, pp. 13-20
- [22] Ishida, N., Inoue, T., Miyahara, M. and Higashitani, K, 2000, Nano bubbles on a hydrophobic surface in water observed by tapping-mode atomic force microscopy, *Langmuir*, vol. 16, pp. 6377-6380.
- [23] Tyrrell, J.W.G. and Attard. P., 2001, Images of nanobubbles on hydrophobic surfaces and their interactions, *Phys. Rev. Lett*, vol. 87, 176104.
- [24] Zhang, X.H. and Khan, A., 2007, "A nanoscale gas state", *Phys. Rev. Lett*, Vol. 98, 136101.
- [25] Angrawal, A., Park, J., Ryu, D.Y., Hammond, P.T., Russel, T.P. and Mckinley, G.H., 2005, "Controlling the location and spatial extent of nanobubbles using hydrophobically nanopatterned surfaces", *Nano Letters*, Vol. 5, pp. 1751-1756
- [26]Yang, S., Dammer, S.M., Bermond, N., Zandvliet, H.J.W., Kooij, E.S. and Lohse, D., 2007, "Charaterization of nanobubbles on hydrophobic surfaces in water", *Langmuir*, Vol. 23, pp. 7072-7077