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EXPERIMENTAL STUDY OF THE BOILING PERFORMANCE ENHANCEMENT USING SURFACE MODIFICATION

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ABSTRACT

CHF(critical heat flux) and boiling heat transfer were key parameters to decide the operation condition of the thermal-hydraulic system. In this study, we have tried the surface modification to enhance the boiling performance (CHF and boiling heat transfer rate) on the metal and the silicon. Anodization was applied to modify the metal surface, and MEMS technique was used to modify the silicon surface. With pool boiling experiment, we found that the peak of CHF enhancement on the modified surfaces was two times higher than bare surfaces. To get the boiling curve during the experiment, the boiling heat transfer could also be increased on the modified surface which has the hydrophilic and the hydrophobic materials. As a previously well-known the effect of surface condition, the wettability has a powerful effect to the boiling condition. So by using the different wettability materials, the boiling condition is enhanced. To investigate these controlled parameters of the modified surface, we measured static contact angle, observed the SEM image of modified surfaces.

INTRODUCTION

The problem of cooling has become increasingly critical in the nuclear industry. The most effective way of cooling a 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. However, boiling heat transfer has an inherent limitation: the critical heat flux (CHF). The CHF is the maximum heat flux that occurs when boiling heat transfer has a high cooling efficiency. When a surface reaches the CHF, it becomes coated with a vapor film that interferes with the contact between the surface and the ambient liquid, and decreases the heat transfer efficiency. The system temperature increases and failure occurs if the temperature exceeds the limits of the system's constituent materials. For this reason, every system incorporates a safety margin by running at a heat flux much lower than the CHF; however, this approach reduces the system efficiency. This compromise between safety and efficiency is an important problem in the nuclear industry.

State of art for increasing CHF under pool boiling

Nanofluids

Nanofluids are engineered heat transfer fluids consisting of nanosized particles (nanoparticles) dispersed in a base liquid. These fluids have been studied in various fields of thermal engineering since Choi [1] began research with nanosized particles well dispersed in engineered fluid. You et al. [2] found that adding tiny amounts (less than 0.001% by volume) of alumina nanoparticles to a conventional cooling liquid could significantly increase the CHF up to 200%. Kim et al. [3] conducted pool boiling CHF experiments with pure water on a heater fouled by nanoparticles resulting from pre-boiling in a nanofluid. This produced the interesting result that the same magnitude of significant CHF increase in a nanofluid was observed for the nanoparticle-fouled surface submerged even in pure water. This suggested that the CHF increase in nanofluids is caused by the altered surface characteristics due to the surface deposition of nanoparticles during nanofluid boiling. The same results were recently obtained by Golubovic et al. [4]. This suggests that interfacial parameter improvements due to nanoparticle fouling could be a key factor in the significant increase in the nanofluid CHF.

Kim et al. [5] reported that nanoparticle-fouled surfaces had significantly greater wettability measured by a reduction in the static contact angle. They based their suggestion on a review of the prevalent CHF theories, stating that the improved wettability caused by the nanoparticle layer could predict CHF enhancement. Liu and Liao [6] and Coursey and Kim [7] performed pool boiling experiments with water-based and alcohol-based nanofluids on a plain heated surface and found that the deposition of the nanoparticles on the boiling surface changed the microstructure and the physicochemical properties of the surface. Such changes in the heat transfer surface significantly influenced the boiling phenomena by changing key parameters such as nucleation site density, bubble departure diameter, bubble frequency, and evaporation of the micro and macrolayer beneath the growing bubbles. However, a heat transfer system using nanofluid raises questions about the permanence of the nanofluid because of the change in volume concentration of the working fluid (nanofluid).

Surface modification

Some research groups have recently developed an artificial structure to resemble the structure generated by boiling in a nanofluid. Kim et al. [8] experimented with the CHF enhancement of an artificial surface to imitate the nanoparticle deposition layer formed after pool boiling in a nanofluid. Flat, micro-, nano- and micro/nanosurfaces formed on a silicon wafer using microelectromechanical techniques had different CHF values. The effects of surface structure and surface wettability were dominant factors in the CHF enhancement. In addition, the spreading effect of nanosized rods on the micro/nanosurface included the wettability effect and produced a greater CHF value than the other factors. In a more realistic application, Zircaloy-4 was used as the test material for the pool boiling CHF experiment in this study; this material is often used as the cladding of nuclear fuel rods. However, the question remained as to whether the CHF on Zircaloy-4 surface could be enhanced by some method, perhaps using non-traditional metals such as copper or stainless steel. In this study, various methods of surface treating Zircaloy-4 were tested to achieve a more wettable surface. There are several ways to increase surface wettability. Because the wetting phenomenon is governed by the surface energy and surface morphology, modification methods can be classified into two corresponding categories. Thermal oxidation is a simple easy method of modifying the surface energy to increase the wettability because metal oxide is more hydrophilic than pure metal. Many researchers in the 1980s used thermal oxidation to make hydrophilic heater surfaces [9]. However, simple thermal oxidation has certain limitations and is capable of producing highly wettable surfaces. Ultraviolet radiation is another method that has been used to modify the surface energy of materials. High-energy radiation changes the surface energy of the substrate and can even produce a super hydrophilic (contact angle < 5°) state. Researchers have recently used this method to increase wettability and confirm the increase of heat flux [10]. However, this method is not very practical for industrial use because the effect is not permanent.

State of art for increasing boiling heat transfer under pool boiling

The enhancement of nucleate boiling heat transfer enables the equipment to be operated in a higher performance under the same limitation. So 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 nanofluids experiment which exhibits an incredible enhancement of CHF when used as a working fluid in pool boiling. It has been proved that the outstanding CHF enhancement is due to the changed surface (Kim et at., 2006). In this consideration, many researches were contributed to improving boiling condition with various methods such as coating the porous media [11], making the surface micro-structure [12, 13] or changing the roughness [14, 15]. 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 higher pool boiling CHF [16]. From this point of view, a new CHF modeling is suggested with consideration of the contact angle of the heating surface [16]. The wettability significantly affects not only the CHF but also the nucleate boiling heat transfer. Recent studies have shown that the nucleation site density, which is a dominant factor on nucleate heat transfer, is related with the surface wettability. 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 [17]. 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).

We would introduce various methods of boiling performance enhancement through the previous studies of two phase flow laboratory of POSTECH and propose the further way of boiling group to have most boiling performance.

NOMENCLATURE

q''	W/m^2	heat flux
h	[kJ/kg]	latent heat
g	[m/s^2]	gravity
Special characters		
σ	[N/m]	Surface tension
ρ	[m]	density
Subscripts		
l		Liquid
g		Vapor

EXPERIMENTAL PREPARATION

This study was conducted by two experimental facilities which were used for kinds of experimental purposes. For CHF enhancement, we took a zircaloy-4 by the method of surface modification as anodization on metal surface, so we took the heat flux on heating surface like thermal conduction heating. And for boiling heat transfer enhancement, we took a silicon wafer by the method of MEMS technique as Teflon dot on silicon surface.

Pool boiling experimental facility of metallic heater

The same experimental facility is used in this paper as that used by reference [18, 26].

Pool boiling experimental facility of silicon heater

The same experimental facility is used in this paper as that used by reference [8, 19].

Test specimen of silicon surface

To facilitate both CHF test and surface modification, we embedded a thin film heater on one side of a silicon wafer and created artificial surfaces on the other side of the wafer by using the MEMS technique. Although the fabrication processes of the film heater side and surface modification were conducted simultaneously, we will describe them separately for convenience of explanation.

The test heater is a rectangular silicon wafer plate (Fig.1). The substrate silicon plate is 25 mm x 20 mm, and has a SiO2 layer for electrical insulation on both the top and bottom. Because Joule heating was selected for this experiment, insulation layers were needed to exclude the possibility of electrical interference on both modified surface structures and of hydrodynamics on the top. For heating part, a titanium thin film of about 1000 Å thickness was layered on the bottom of the substrate using an E-beam evaporator. The completed titanium film heater has 'H' shape. The center bar of 'H' is 15 mm x 10 mm with a vertical bar at each end. The center bar is the main heat generating area, and we confirmed by simple calculation that it generates more than 99 % of the total heat. The vertical bars at ends are used for wire connection. Because it is impossible to use lead soldering to connect copper wires to a titanium film, an additional gold (Au) film (1500 Å thickness) was deposited on each vertical bar using an E-beam evaporator. With lead soldering and Au film on the vertical bars, their resistance becomes smaller than that of the center bar and guarantees that heat generation is focused in the center bar.

To increase the CHF under pool boiling, the surface of test specimen was modified (Fig. 2). The more detailed explanation could be referred by the previous study [8]. And to increase the boiling heat transfer ability under pool boiling, the surface of test specimen was conducted by Teflon dot method like the previous study [19] and Fig. 3. The surface characteristic of silicon heater was presented on the reference [8, 19].

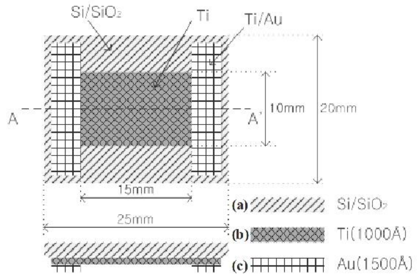

Figure 1 Test heater fabrication of silicon surface [8].

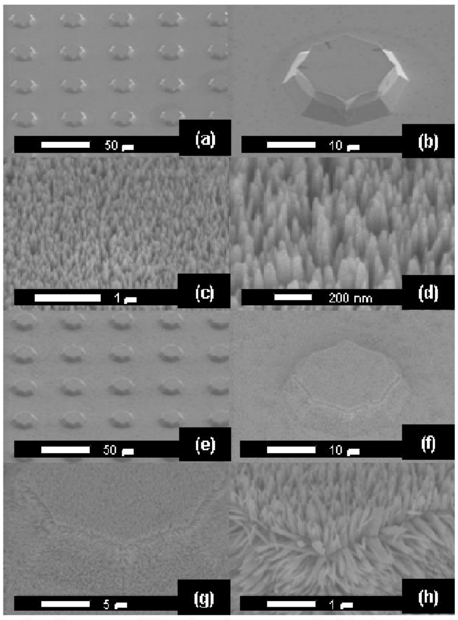

Figure 2 Surface modification by MEMS fabrication on silicon oxide wafer : (a) and (b) micro structured; (c) and (d) nano structured; (e),(f),(g) and (h) micro-nano structured [8].

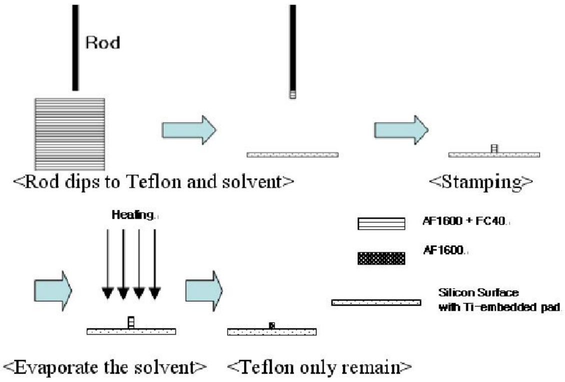

Figure 3 The process makes the hydrophobic dot on silicon surface [19].

Test specimen of metallic surface

To conduct the application, we choose the material of heater as metal. To easily modify the metal surface, anodic oxidation method was taken. It is an electrochemical fabrication technique in which an external applied electric potential is used to enhance the chemical reaction at the interface between an electrolyte and the target material. The reaction rate can be very easily controlled by adjusting the applied voltage; this is as sensitive as a 108-fold increase of the reaction rate per volt of increased potential [20]. Varying the other parameters such as current density, type of electrolyte, and acid strength also affect the reaction and result in a different end product. The final product of anodic oxidation is a generally a thick oxide layer. However in certain materials and under certain conditions, unique ordered nanostructures occur instead of randomly formed oxide structures. Anodic aluminum oxide (AAO) [21] and titania nanotubes [22] are two well-known examples. Because of their nanoporous surface morphologies and the chemical nature of the metal oxide, these examples are very hydrophilic. Anodic oxidation of zircaloy has already been attempted and the results have been observed by microscopy [23]. However, no ordered nanostructures have been found and few studies have attempted to measure the resulting wetting properties. Ever since titania nanotubes were first reported in the early 2000s, we have followed the progress of related research very closely because both titanium and zirconium are in the same chemical group in the periodic table, and similar forms can be expected for zirconium alloy. Zirconia nanotubes were indeed subsequently reported by Lee et al. [24]. After in-depth study of the preceding results, we finally succeeded in fabricating zircaloy nanotubes. We then conducted wettability studies on these nanotubes as well as the associated micro/nano -scale mountain-like structures that were subsequently discovered. Rectangular zircaloy-4 plates (20 × 25 × 0.7 mm) were used as test samples. They were mechanically polished with #1200 silicon carbide abrasive to remove impurities and produce a uniform surface. The polished samples were cleaned with a 1:1 mixture of acetone and methanol in an ultrasonic bath. After rinsing in deionized water, the samples were completely dried [25]. The detail process of anodization was published on reference [26]. The surface characteristic of metallic heater was also presented on the reference [26]. The SEM image of anodic oxidized zircaloy-4 heater was showed by Fig. 4.

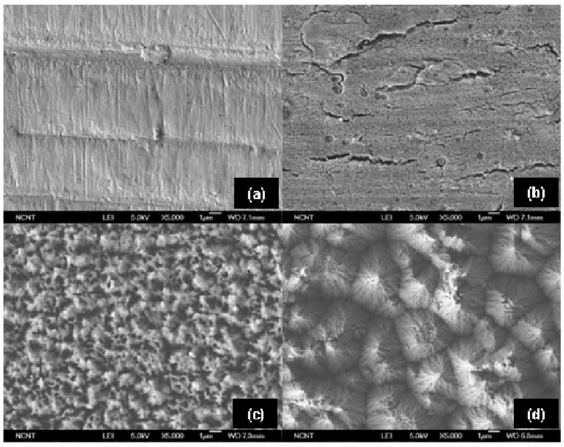

Figure 4 Surface of modified zircaloy-4: (a) bare; (b) oxidized meaningless structure; (c) nano structured; (d) micro-nano structured [26]

EXPERIMENTAL RESULTS AND DISCUSSION

All experiments were conducted under saturated pool boiling at atmosphere pressure (101.3 kPa) and heat flux was applied from the convective heat transfer regime to CHF. The uncertainties analyses of all data were referred by previous studies. We only presented the boiling curve, CHF and surface SEM images and referred the previous studies about quantitative data of surface characteristics; e.g. static contact angle, roughness and spreading abilities.

Enhancement of CHF under pool boiling

Figure 5 shows the boiling curve of modified surface of silicon oxide. Every modified surface showed noticeable CHF enhancement compared with the reference bare surface. The average CHF was 1121 kW/m^2 for bare, 1652 kW/m^2 for micro structured, 2003 kW/m^2 for nano structured and 2326 kW/m^2 for micro-nano structured. Differences in CHF estimates on samples of the same surface differed by < 15%. The micro-nano structured surface showed the most outstanding enhancement (107 % increase in CHF compared to the bare surface), followed by the nano structured (79 % increase) and micro structured surfaces (47 % increase). The array of micro-scaled posts caused CHF enhancement of kW/m^2 (from bare surface to micro structured surface) and 323 kW/m^2 (from nano structured surface to micro-nano structured surface). The nanorods caused a CHF enhancement of 882 kW/m^2 (from bare surface to nano structured surface) and 674 kW/m^2 (from micro structured surface to micro-nano structured surface). The test results were compared with the well known CHF models of Zuber [27]. Zuber's correlation for an upward-facing horizontal flat plate is

$$q''_{CHF\_Zuber} = 0.131\, h_{lg}\, \rho_{lg}^{0.5}[\sigma g(\rho_l - \rho_g)]^{0.25} \tag{1}$$

It predicts 1108 kW/m^2 and agrees quite well with the CHF value (1121 kW/m^2) of the flat plate sample (bare surface). Because Zuber's correlation does not consider the effects of surface modification on test samples, it cannot explain the CHF enhancement of the other test samples.

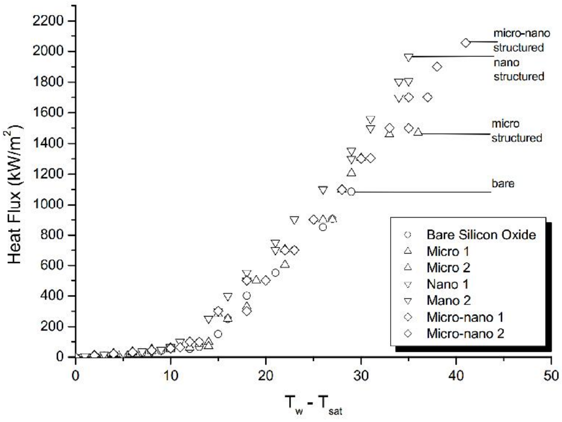

Figure 5 Boiling curves of modified silicon surfaces

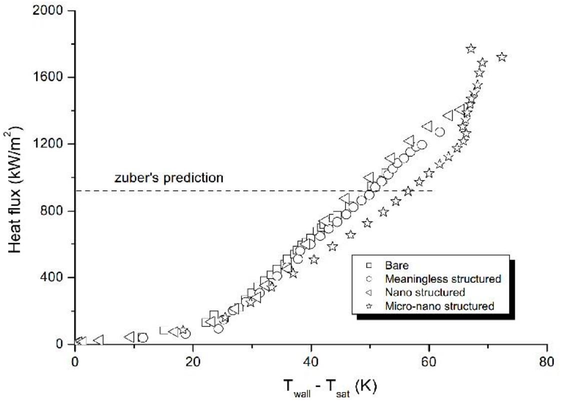

Figure 6 Boiling curves of modified zircaloy-4 surfaces

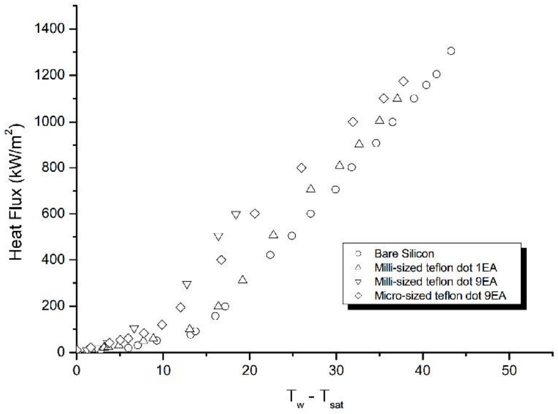

Figure 7 Boiling curves of hydrophobic dot surface

Figure 6 shows the boiling curves of modified zircaloy-4 surfaces. As previously mentioned, nano and micro-nano structured have higher CHF value as bare and meaningless structure. Various parameters could influence CHF directly and indirectly. Among these parameters, wettability and spreading ability of surface influenced CHF enhancement. Surface characteristics of not only modified silicon oxide water, but also modified zircaloy-4 have more wettable and spreadable than each bare surface. Nanotube-like nanostructures can be observed in Fig. 4. These nanostructures aided the liquid spreading ability of the treated zircaloy-4 to some extent and played a role in the CHF enhancement. The same surfaces also contained valley-like microstructures, as shown in Fig. 4. Chen *et al.* [28] reported that microscale structures could enhance capillary wicking and capillary-driven flow. Ishino *et al.* [29] reported that microscale structuress could influence liquid capillary wicking on the surface, and the liquid spreading ability could be characterized by the diameter of the wetted zone that surrounded the liquid droplet. Kim *et al.* [8] reported that nanostructures could help the liquid spreading and delay the CHF. In addition, they reported that the combination of nano- and microstructures had the best liquid spreading ability and the best enhanced CHF. The micro/nano multiscale structures that facilitated the liquid spreading on the heated surface were like a porous medium.

Enhancement of boiling heat transfer under pool boiling

Figure 7 shows the boiling curves in the experiment with the bare silicon surface, milli-dot, and micro-dots on the silicon. In the view of nucleate heat transfer coefficient, the sample is divided into the silicon surface with the hydrophobic dot and without it. The result shows that the surface with the hydrophobic dot has a better nucleate heat transfer than without, being considered that the hydrophobic dot makes early ONB as previously discussed. The high nucleate heat transfer is highlighted with the micro-dots surface, because a dot smaller than the milli-dot makes a higher bubble frequency at a given heat flux and thus the total volume of bubbles on the micro-dots will be larger than that on milli-dot when all hydrophobic dots are activated on the same Teflon total area. Actually the difference of nucleate heat transfer between the milli-dot on silicon and bare silicon is not distinctively large because the ratio of the total area of the hydrophobic material to total heating area is not so much. From this observation, it is evident that the second experiment with the milli-dot should be needed. The milli-dot on the silicon gives a lower CHF than on the bare silicon. It is conjectured that the center dot may influence on CHF. On the other hand, the CHF data from the micro-dots are scattered. CHF's from some micro-dot experiments are comparable to that of the bare silicon, while the other micro-dot experiment gives almost the same CHF as that from the milli-dot experiment. The result is most likely to come from the uncertainty in producing micro-dots which can affect the bubble inception without changing CHF significantly. Fig 6 shot by a high speed camera shows bubble inception on the bare silicon near the onset of nucleate boiling (ONB, wall temperature is near 115oC here). A bubble grows on a cavity, and then the bubble smoothly departs from the cavity after bubble elongated in a little while.

Figure 8-(a) shots by a high speed camera shows bubble inception on the bare silicon near the onset of nucleate boiling (ONB, wall temperature is near 115°C here). A bubble grows on a cavity, and then the bubble smoothly departs from the cavity after bubble elongated in a little while. Figure 8-(b) demonstrates a bubble behavior on the Teflon milli-dot. The bubble on the Teflon milli-dot appears earlier than on the bare silicon when the wall temperature gets near 101~3°C. The first initiating bubble is generated on the Teflon dot, and its size seems to be related with the dot size. The heat flux at ONB on the Teflon milli dot is so small that bubble frequency is very low. The other feature of bubble on the hydrophobic dot is that the bubble sustains saturated state for a certain time and then departs suddenly. The saturated bubble state means that bubble doesn't grow and doesn't depart from the Teflon dot either. The sudden departure is the phenomenon that occurs when the bubble buoyancy force gets over the strong force of triple line on the different surface border with different fluid state (It means the boundary of bubble: liquid and vapor). Because the size of the bubble incepted on the Teflon dot is seized by the

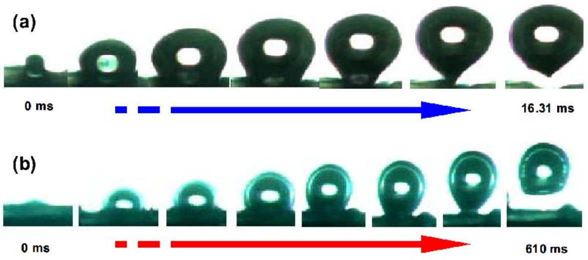

Figure 8 Bubble growth and departure: (a) bare silicon oxide; (b) Teflon mili-dot

teflon dot size, the bubble frequency is only dependent on the heat flux. It is evident that the more heat is supplied, the higher bubble frequency is made.

CONCLUSION

The thermal system using boiling heat transfer has the limitation of CHF phenomenon and need the more efficient heat transfer performance. As previous researches, important parameters of CHF and boiling heat transfer have been known by surface effect. We propose the surface wettability and spreadibility related CHF enhancement, and the different surface wettability; e.g. hydrophilic and hydrophobic, related boiling heat transfer. If we propose the proper surface which was well combined by these surface properties, boiling performance on proposed surface was expected to work well at real field. Furthermore, we have a plan to design the proper surface which has a good boiling performance increasing both CHF and boiling heat transfer.

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