Enhanced Boiling Heat Transfer from a Novel Nanodendritic Micro-porous Copper Structure

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Abstract

Following licentiate thesis is a summary of the advances made within the research project - Micro- and nano structured surfaces for enhanced boiling heat transfer – which is a collaboration effort between the Division of Applied Thermodynamics and Refrigeration and the Division of Materials Chemistry at the Royal Institute of Technology (KTH).

The main objectives with this research project has been to: develop methods for producing highly efficient boiling surfaces with well defined micro- and nano-structured porous surfaces by the use of micro- and nano-manufacturing techniques. This objective has been achieved and the result is a novel micro-porous surface structure comprising dendritically ordered nano-particles of copper. The structure was fabricated by a high-current-density electrodeposition process, in which the evolution of hydrogen bubbles serve as a dynamic masking template to the growth of the dendritic copper structure. Important variables were identified that affect the production of the structure and its features, such as surface orientation during electrodeposition, pressure and temperature of electrolyte, and a final heat treatment of the surface under reduced atmosphere, all of which have previously not been reported on.

Experimental tests have been conducted in a widely used refrigerant, R134a, where the micro-porous structure was shown to enhance the boiling performance of a copper surface over 15 times compared to a regular copper surface. The boiling characteristics of the structure were found to be dependent on controllable surface characteristics. The remarkably good boiling performance of the novel micro-porous enhancement structure has been attributed to its high porosity (~94%), a dendritically formed and exceptionally large surface area, and to a high density of well suited vapor escape channels (>50 per mm²).

A patent application, intended to protect the enhancement structure and its fabrication method, was submitted to the Swedish patent authorities (PRV) on March 1st, 2006.
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1 Introduction

1.1 Why Enhanced Boiling?

In an attempt to answer this question, a number of benefits associated with enhanced boiling technology are here presented.

**Energy Efficiency** - as engineers, we are often occupied with energy efficiency, and those of us, who are active in the field of heat pumping and refrigeration technologies, are no different. We all know, that a reduction of the temperature difference in an evaporator or a condenser by 1 °C results in an improved energy efficiency of somewhere in the range of 2-4%, based on the Carnot efficiency. Considering that a five degree reduction, could save us up to 20% in energy consumption and that billions of dollars are spent annually to power heat pumping machines, no wonder that there is an intense hunt for reductions in “delta Ts” (temperature difference). In enhanced boiling heat transfer, the temperature differences in evaporators can be reduced through various ways of improving the mechanisms of heat transfer.

**Miniaturization** - who remembers the days when a cell phone could not fit into your pocket or when computers were the size of buildings? Space efficiency, or miniaturization, is one of the strongest incentives for technological development. Small components that can perform the same function as larger ones, are not only minimally invasive, but tend to come with a host of benefits apart from the obvious advantage of more efficient use of resources. If we can exchange the same amount of heat in a third of the space, what can we not do with the space freed up? Perhaps we could add new functions that formerly would not fit, lower the weight of the product and thereby finding new applications, etc. The list is endless. Miniaturization may also lead to favorable scaling laws when forces that scale with a low power become more prominent in the micro domain, e.g., surface tension forces become increasingly important than gravity forces in a narrower capillary. By the use of enhanced boiling, the same amount of heat can be transferred over a smaller area.
Reliability and Safety - for cooling of constantly increasing power dissipation from CPU chips and microprocessors, new and innovative heat transfer surfaces are continuously being developed and tested. Direct liquid cooling, involving boiling heat transfer, is one of the most effective ways to maintain a low component temperature and thereby assure a reliable operation of the unit. At very high heat fluxes, a regular boiling surface will transition into the film boiling regime (the so called critical heat flux, CHF) and thereby experience a rapid temperature increase of several hundred degrees. This would result in a breakdown of the electronic component, but with enhanced boiling techniques, the CHF can be increased and thus ensure continuous cooling of the component. As we are acutely aware of, reliable and effective cooling in nuclear power reactors is of highest priority, where transition to film boiling can have devastating consequences. Here, enhanced boiling techniques may increase the safety margin considerably.

Economic Efficiency – As alluded to above, the economic benefits of enhanced boiling can be very compelling and extend beyond the cost side of the business equation. There is a monetary value to all the advantages listed above; better energy efficiency, a more reliable product, new functionality, improved mobility, etc. These competitive advantages are often worth far more than the cost savings that would come from reduced use of material for a smaller heat exchanger.

As a result of these benefits, enhanced boiling technologies have made important inroads into the refrigeration and air-conditioning industries, the oil, gas and chemical processing industries, the microelectronics industry, the space industry, and several others.

1.2 A brief orientation

There are several ways in which to improve the boiling. The main distinction is usually made between passive and active techniques. To augment boiling, passive techniques employ special boiling surface geometries, or fluid additives, such as roughening of the heat transfer surface, pitting the surface with corrosive chemical, integral fins or rolling, knurling integral finned tubes, coating the surface with a porous layer, etc. Active techniques need external power, such as electric or acoustic fields and surface vibration. Most of these active techniques have proven to be difficult to realize outside the laboratory or to justify economically.
During the past few decades, a considerable amount of articles and research reports have been published concerning the issues associated with nucleate boiling. For a comprehensive literature review of the research within the field of enhanced boiling, the reader is referred to a literature survey by Furberg (2006)\(^1\).

### 1.3 Thesis at hand

The work presented here has its focus on passively enhanced pool boiling. The research project is a unique collaboration effort between the Division of Applied Thermodynamics and Refrigeration and the Division of Materials Chemistry at the Royal Institute of Technology, in which research and competences from two different fields of science are combined and applied. Recent discoveries within both fields of expertise have made valuable synergies possible by which both sciences will benefit from combined research.

First, recent developments within nano- and micro-technologies have made possible the creation of well structured surfaces down to the nanoscale, by template techniques and by a novel electrochemical deposition process. Secondly, new findings have shown the importance of nanosized inhomogeneities of the surface for the boiling performance of the surface.

The combination of these developments has opened up the possibility to explore the connection between the micro- or (nano-)scopic topology of the surface and the macroscopic boiling performance, thereby allowing the modeling of the bubble nucleation process and optimization of boiling surfaces.

#### 1.3.1 Objective

The overall research objectives of the doctoral thesis project are:

- To develop methods for producing highly efficient boiling surfaces with well defined micro- and nano-structured porous surfaces by use of micro- and nano-manufacturing techniques – this objective has been achieved within the scope of this licentiate thesis.

- To study the microscopic nucleation behavior and the macroscopic heat transfer performance of the developed surface and relate the behavior to the structure of the surface – this objective has been achieved within the scope of this licentiate thesis.
To use the developed models and manufacturing techniques to produce highly efficient boiling surfaces suitable for exploitation in commercial equipment – this objective has been achieved within the scope of this licentiate thesis, but due to the patent applications that is currently under review and ongoing and confidential industry sponsored application research, these findings will not be presented here.

To develop models for predicting the nucleation process and the boiling heat transfer performance of micro- and nano-structured surfaces from a geometric description of the surface and known thermodynamic and transport properties of the fluid and the surface material – this objective is yet to be achieved.

1.3.2 Methods
Experimental methods have been used in developing a highly efficient boiling surface and testing the surface's heat transfer performance. The development of a new surface structure has consisted of testing several different ideas through visual observation of the fabricated surface in a scanning electron microscope (SEM), measuring the various structural and geometrical parameters, such as weight, porosity, pore-density, thickness, etc., and also experimentally measuring the mechanical stability of the surface structure. The boiling performance of the different surface structures was experimentally evaluated in a pool boiling test rig (details presented in section 4).

In enhanced boiling heat transfer, the researcher often has the opportunity to significantly improve the thermal performance of a heat transfer process. This improvement can rarely be obtained through following standard, ordinary design procedures, but rather through much creativity and innovation. Every heat exchanger is a potential candidate for enhanced heat transfer and herein lays much of the fun and the challenge with the research in this field.
1.4 References

2 Advances in Enhanced Boiling

The area of passive pool boiling enhancement technology has been subject to an interesting development during the last couple of years and, to a large extent, advances in the field of micro- and nano-technology have fueled this development. Furthermore, recent progress that has been made in the areas of computational fluid dynamics (or numerical simulations), and measurement & control technology has also been driving the current development within the sphere of pool boiling research.

Here follows a brief summary of recent and significant developments within these research fields and how these advances translate into more efficient boiling surfaces.

2.1 Micro- and nanotechnology

None of us have been able to escape the reports on the emergence of nanotechnology, where materials, structures or devices are given new properties by means of controlled manipulation of their microstructure at the atomic or molecular level. Now, when nano-size building blocks can be arranged into high dimensional structures with controlled interfaces, new, and very promising surface structures can be manufactured for enhanced boiling applications.

Honda et al. (2002) have published a very interesting study on the effects of micro-pin fins and submicron-scale roughness on the boiling heat transfer from a silicon chip in the dielectric liquid FC-72 ($T_{sat}=56 \, ^\circ\text{C}$ at 1 atm). The enhanced structure was fabricated by the use of dry etching technique and the submicron-scale roughness was fabricated by depositing a thin layer of SiO$_2$ on the silicon surface by sputtering and followed by a wet etching step. The surface, consisting of pin-fins (50x50x60µm,width;thickness;height), was tested with (chip EPF) and without (chip PF) submicron-scale roughness was and compared to two plain silicon surfaces, with (chip E) and without (chip S) submicron-scale roughness, see Figure 1. The fin pitch was 100 µm.
The tests were conducted with vertical and horizontal heater orientation. As illustrated in Figure 2 (left), the micro pin-fin chips were effective in enhancing heat transfer, boiling incipience and CHF (1.8-2.3 times). The most interesting result was that submicron-scale roughness further improved the enhancement for both the plain and the pin-fin chips. Hence, it was confirmed that nano-scale surface features affect the boiling heat transfer mechanisms. Furthermore, a high-speed video study of the boiling phenomena revealed that small amount of vapor was left within the gap between the pin fins when a growing bubble left the surface, which suggested that the evaporation that occurred within the small gap was responsible for the sharp increase in the heat flux with increasing wall superheat.

Wei and Honda (2003)² also studied the effects of fin geometry of these micron-sized pin-fins on silicon chips (Figure 1), fabricated by dry etching. The square fins were ranging from 30x60µm to 50x270µm (thickness x height) with a fin pitch that was twice the fin thickness for all chips. The micro-pin-finned chips showed a considerable boiling heat transfer enhancement and increase in CHF, compared to a smooth chip, see Figure 2 (right). In general, the larger sized fins showed the best per-
formance. The effects of dissolved gas were also tested and, not surprisingly, it was concluded that dissolved gas mainly has a positive effect in the low flux region. A vertically mounted chip performed better than a horizontally orientated chip in the low flux region, the same in the medium heat flux region, but had a lower CHF.

The boiling behavior of the micro-pin-finned chips was characterized by a very sharp increase in heat flux, $q$, with increasing temperature difference, $\Delta T_{sat}$, as may be seen in Figure 2. Based on high speed photography, it is believed that the bubble nucleation occur mainly at the fin flank. With a uniform surface roughness for all fins, a regular geometry and the fact that all micro-pin-fins are completely submerged in the layer of superheated liquid, the condition for bubble nucleation will be satisfied at almost the same $\Delta T_{sat}$ at all parts of the chip surface. Hence, the number of nucleation sites will increase rapidly as $\Delta T_{sat}$ increases, thus resulting in nearly isothermal behavior (a vertical boiling curve).

Akapiev et al. (2003) and Schulz et al. (2005), were the firsts to present a geometrically well-defined microstructure with cylindrical copper whisker, see Figure 3. The fabrication process, illustrated in Figure 4, starts with a polycarbonate foil that is irradiated by heavy ions and thereafter etched to produce a membrane with cylindrical pores, the so-called ion track membrane. The membrane is wrapped around the copper tube that serves as a cathode in the galvanic cell. The subsequent deposition process fills the pores with copper. An array of cylindrically shaped micro pins, with a porosity of 75 to 99%, is created on the tube surface after dissolving the membrane. A simple acetone evaporation experiment showed that the microstructure possesses good capillary characteristics involving a full distribution of acetone over the surface and therefore also seemed as good candidate for an enhanced boiling surface.
Mitrovic and Hartmann (2004)\textsuperscript{5} conducted pool boiling tests of this microstructure with cylindrically shaped micro pins in R141b. As illustrated in Figure 5, the boiling incipient point was lower and the heat transfer coefficient was improved over the whole heat flux range with approximately a factor 2. An interesting observation was that the structure showed an isothermal behavior at varying heat flux, in the fully developed boiling region. This kind of isothermal boiling behavior can also be seen in the micro pin-fins, fabricated on silicone chips, tested by Wei and Honda (2003)\textsuperscript{2}, as presented in Figure 2.

Mitrovic (2005)\textsuperscript{6} speculates that the required properties, of a surface structure with isothermal boiling behavior, must be that of identical elements which are arranged in a mono-pattern on the heating surface. The structure elements (protrusions) must trap vapor after bubble detachment and generate a possibly long three-phase-line (TPL) formed by in-

\begin{figure}[h]
\centering
\includegraphics[width=0.45\textwidth]{figure4.png}
\caption{Fabrication method for cylindrical copper whisker.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=0.45\textwidth]{figure5.png}
\caption{Boiling curve with isothermal behavior.}
\end{figure}
intersection of the vapor-liquid interface with the heating surface, see Figure 6. These properties are also characteristic of the Wei and Honda (2003)\textsuperscript{2} structure with its square pin fins. A longer TPL than the one featured by the cylindrical micro-pin fin structure, would be possible if the structure elements would have a longer cross section, i.e. zig-zag or fractal like, see Figure 6.

Another interesting set of studies of a well-defined micro- and nano-structure have been presented by Ramaswamy et al. (2002)\textsuperscript{7}, Ramaswamy et al. (2003)\textsuperscript{8}, Ghiu and Joshi (2005)\textsuperscript{9}, Ghiu and Joshi (2005)\textsuperscript{10}, Murthy et al (2005)\textsuperscript{11}. The enhanced structure consisted of micro-channels with various channel width, pitch, and height in the range of 65-1000 µm, see Figure 7. Wafer dicing and wet etching was used to fabricate well defined net of interconnected micro-channels on a 10 mm x 10 mm piece of silicon wafer. The resultant structure had pores that communicate the interior of the micro-channels with the liquid pool. The structure was also produced in copper and in quartz employing an automated dicing saw. The structure showed a maximum of five times improvement of the heat transfer coefficient at low heat flux and improvements in the incipience boiling point and the CHF was also found.

Figure 6. (Left-top) Piercing the interface by pins results in a large TPL. (Left-bottom and Right) Cavity formed by neighboring pins. (Right) The von Koch fractal curve as border of the cross-sectional area of a structure element.

Figure 7. Enhanced structure in silicon, copper and quartz.
High-speed visualization studies were made of the pool boiling tests in both FC-72 and PF 5060. The data from these visualizations served as a basis for the development of two different semi-analytical models for boiling from the enhanced structures, where the heat transfer was quantified by predicting the bubble departure diameter, bubble frequency, nucleation site density and dissipated heat flux within an accuracy of ±30% of experimental data.

Launay et al. (2005)\textsuperscript{12} has further modified the micro-porous silicon structure (Figure 7) tested by Ramaswamy et al. (2002)\textsuperscript{7}, with a coating of carbon nanotubes (CNTs) and thereby created a hybrid micro-nano structure for boiling enhancement. Tests were conducted in PF5060 and in deionized water. The surfaces tested were; smooth and roughened silicone surfaces, bare and fully coated with CNTs, a silicon etched pin-fin array as well as a CNT-based pin-fin array, and 3-D structures with and without CNTs. The carbon nanotubes were grown on the silicone oxide overcoat patterned on the surface using a chemical vapor deposition as described by Wei et al. (2002)\textsuperscript{13}, with an inner diameter of the CNTs in the 10-20 nm range and with a pitch of 50-70 nm between individual tubes.

The conventional silicon fin-pin microstructure and the 3D microstructures showed the best performance, but the CNT-enabled structures improved boiling at low superheats. The CNT results might have been worsened due to possible oxide formation between surface and heater (during the high temperature copper vapor deposition process) forming a contact resistance. It is also observed that the boiling curves for the CNT-based structures are similar to boiling curves exhibited by hydrophobic surfaces. That is the surface heat flux increases monotonically with superheat, eventually merging with the film boiling curve at the high superheat level. This research shows many similarities with that of Honda et al. (2002)\textsuperscript{1} in that the single and combined effects of micro-
and nano-features are studied. An interesting comparison is that Honda et al. (2002) recorded an enhancing effect from the nano-roughness at all heat fluxes and not only at low heat flux.

Vemuri and Kim (2005) reported on pool boiling experiments from a nano-porous surface in FC-72. A 30% reduction in temperature superheat required for boiling incipience was found. The pore size of the structure was in the 50-250 nm range and the thickness of the structure was 70µm, see Figure 9. The structure, an aluminum membrane filter produced by Whatman Inc. probably aimed for bio-separation applications, was glued to the heated surface. These findings further indicate that even nano-sized pores may trap vapor patches and thereby lower the incipience boiling point.

2.2 New measurement technologies

As we have seen, the boiling is a multi-scale dependent phenomenon where the heat transfer mechanisms are affected by surface features on both macro-, micro- and nano-level. Recent advancements in measurement technologies have made it possible to study the boiling phenomena with very high spatial and temporal resolution. Typically these measurements are performed on idealized flat surfaces, but deepened understanding of the boiling phenomena aids and guides the development of enhanced boiling structures.

Theofanous et al. (2002) in a two part paper, visualized the thermal patterns on nanoscopically smooth surfaces (heaters) using an advanced high-speed and high-resolution infrared camera, see Figure 10. The heater and boiling surface was fabricated by vapor-depositing sub-micron metallic films and characterized with atomic force microscopy,
scanning electron microscopy, and X-ray diffraction spectroscopy. The data show a stark difference between fresh and aged heaters with increased nucleation site density with aged heaters. The origin, evolution and dynamics of the heater dry-out process is identified quantitatively and captured in action.

The authors suggest that, in contrast to the cavity theory of heterogeneous nucleation that requires a presence of gas bubbles entrapped in the heater surface microscopic cavities, nano-scale imperfections and defects present on the heater are sufficient to initiate the heterogeneous nucleation. The data also show a direct correlation between CHF and nucleation site density, which was increasing with aged heaters. Hence, micro-structuring may improve the heaters CHF performance.

Demiray et al. (2004) reports on a new experimental heater configuration of 10x10 micro-heaters, each with 100µm resolution, see Figure 11 (left), allowing direct heat transfer measurements under nucleating bub-
bles, combined with high-speed digital video imaging. Each heater in the array was kept at constant temperature by individual feedback circuits. During these isothermal conditions it was found that the bubble gains most of its energy from the superheated liquid layer surrounding the bubble, rather than the micro-layer between the bubble and the surface or the contact line. Overall, transient conduction/micro convection, during liquid rewetting of the surface, accounted for most of the wall heat transfer.

Myers et al. (2005)\textsuperscript{18}, as an extension of the work by Demiray et al. (2004)\textsuperscript{17}, reports on experiments where the time and space resolved heat flux and temperature distributions under nucleating bubbles on a constant heat flux surface were obtained. The data indicated that most of the energy required for bubble growth came from the superheated liquid layer around the bubble and only 23\% came from micro-layer evaporation and contact line heat transfer, hence in some ways contradictory to the classic micro-layer theory presented by Cooper and Lloyd (1963)\textsuperscript{19} among others. Overall, the dominant heat transfer mechanism was transient conduction into the liquid during bubble departure. Bubble coalescence only had a small impact on the heat transfer rate (bubble coalescence results in a larger bubble which triggers the bubble to departure quicker, hence increasing the departure frequency).

Li and Peterson (2005)\textsuperscript{20} have evaluated bubble incipience and dynamics on a micro-heater 100x100 µm, with and without a manufactured defect (nucleation trigger). High-speed videography was used to visualize the nucleation behavior. The results show that explosive boiling can occur on a smooth surface independent of heating rate. Dissolved gas and surface roughness are shown to have a significant influence on the incipience temperature and bubble behavior. One conclusion from the study is that a classical kinetics of boiling approach can be used to explain boiling incipience in principle.
Höhmann and Stephan (2002)\textsuperscript{21} used temperature sensitive liquid crystals (TLC) and two high-speed digital cameras in conjunction with a microscope to investigate the heat transfer at an evaporating liquid meniscus (not nucleate boiling), with a resolution of less than 1 \( \mu \text{m} \). Their setup enabled two-dimensional measurements of the temperature distribution underneath the evaporating meniscus meanwhile the shape of the meniscus, including the apparent contact angle between fluid and wall, were determined by backlight profiling. The data obtained, showed high heat flux in the micro-region of the meniscus, see Figure 12.

In a follow up article, Stephan and Kern (2004)\textsuperscript{22} investigated, theoretically and experimentally, the heat and mass transfer phenomena in nucleate boiling of both pure substances and binary mixtures. Micro-scale phenomena were recognized as being important for the understanding and prediction of macroscopic heat transfer. Modeling strategies presented included micro-scale heat transfer and were able to accurately predict the heat transfer coefficient. Heat transfer was found to be governed by one-dimensional heat conduction normal to the wall, the molecular interfacial phase change resistance, and intermolecular forces of adsorption. Mass transfer is induced by capillary forces and an evaporative induced flow, see Figure 13. On the macro-scale, heat transfer is governed by transient conduction within the wall and the thermal boundary layer as well as evaporation at the phase interface of the vapor bubbles.
Triggered by findings by Yaddanapudi and Kim (2001), who studied single bubbles growing on a constant temperature heater and who found that over 40% of the latent energy came from heat transferred through the bubble dome (the rest, 60% through the liquid micro-layer), Liao et al. (2004) developed a physical model for early stage bubble growth in saturated nucleate boiling. It was found that the thin unsteady thermal boundary layer near the rapidly growing bubble allows for significant amount of heat flux from the bulk liquid to the vapor dome, which in some cases can be larger than the heat transfer from the micro-layer, see Figure 14. The findings are therefore in general agreement with the experimental results presented by Yaddanapudi and Kim (2001).
2.3 Numerical simulations

To have a global mechanistic model for boiling heat transfer, it is imperative to sub-models that represent the underlying physics of the subprocesses correctly. Boiling heat transfer is a function of a multitude of interrelated sub-processes such as: bubble dynamics, fractional area of liquid-solid contacts and their duration, mechanisms of heat transfer, interfacial instabilities, and interfacial mass and heat transfer as well as other independent variables such as; heater size, shape, thickness and surface condition, heating method, liquid temperature, systems pressure, liquid and vapor thermophysical properties, heater confinement, and flow conditions.

Since these sub-processes are interrelated, the global model will only work if the processes are modeled simultaneously, and because of the profound complexities involved, numerical computer simulations are necessary. Currently, with the rapid improvement in the numerical capabilities and processing speeds of computational fluid dynamics, we are a step closer to the objective of reaching a complete understanding of boiling heat transfer.

Dhir (2001), in a comprehensive article, presents two-dimensional numerical simulations of both pool nucleate and film boiling at normal earth gravity and microgravity conditions. Complete solutions to the conservation equations of mass, momentum, and energy for liquid and vapor phases at and near a heated surface are presented. Both the macro- and the micro region of the growing bubble was simulated, see Figure 15 (left).

![Figure 15. (Left.) Macro- and microregions used in numerical simulations. (Right) Bubble growth pattern.](image-url)
The simulations were validated with experimental results performed under well defined conditions, where the heat transfer rates, bubble release pattern, bubble shape and the interface growth rate were measured. The results from the simulations were remarkably accurate and the bubble dynamics closely resembling the experimental results, see Figure 15 (right). About 20% of the total heat transfer came from micro-layer evaporation. The simulations showed accurate results for nucleate boiling pertaining to the effects of; wall superheat, contact angle, liquid subcooling, level of gravity, heat transfer rates, bubble merger and formation of vapor columns, see Figure 16.

Shin et al (2005) performed a full direct numerical simulation of nucleate boiling in a fully three-dimensional geometry using the level contour reconstruction method, which was developed by Shin and Juric (2002). The effect of nucleation site density and the interaction with other bubbles were included in the model and it was found that the bubble’s effect on neighboring bubbles was very important for predicting the proper relationship between heat flux and the wall superheat in a realistic surface. For simplicity reasons, the important micro-layer evaporation and contact line dynamics beneath the bubble was neglected. The contact region was approximated as fixed in space, which can be seen in Figure 17. The three-dimensional simulations, including the effect of neighboring bubbles, showed better agreement with some known pool boiling correlations, than two-dimensional simulations did.
2.4 References


Section B: Beam Interactions with Materials and Atoms, 236, pp. 254-258.


3 Novel Nano-structured Micro-porous Surface

Since one of the objectives with this research project has been to: *develop methods for producing highly efficient boiling surfaces with well defined micro- and nano-structured porous surfaces by use of micro- and nano-manufacturing techniques*, we have had to put on our thinking caps. The result is a novel micro-porous surface structure comprising dendritically ordered nano-particles of copper. The structure was fabricated by a high-current-density electrodeposition process, in which the evolution of hydrogen bubbles serve as a dynamic masking template to the growth of the dendritic copper structure.

The micro-porous structure has, in experimental tests, proven to considerably enhance the boiling performance of a copper surface. A patent application, intended to protect the enhancement structure and its fabrication method, has been submitted to the Swedish patent authorities (PRV, application date: 3/3/06, application number: 0600475-8, authors: Furberg, R., Li, S., Mamoun, M., Palm, B., Toprak, M.).

This novel boiling enhancement structure may be used in all types of heat exchangers, such as plate heat exchangers, inside or outside tubes in tube-in-shell heat exchanger, hot surfaces in electronics cooling, the evaporating side of heat pipes, refrigeration equipment, air conditioning equipment and heat pumping equipment, thermosyphons, high-efficiency evaporators, in the cooling channels inside water cooled combustion engines and the like.
3.1 Overview of Fabrication Technique

Electrodeposition has been recognized, by for instance Xiao et al (2004)\(^1\), as a suitable process to build and modify three-dimensional structures. Hydrogen evolution during electrodeposition is usually suppressed, since it causes low current efficiency and decreases the density of the deposited metal layer. However, in this work, the hydrogen bubble evolution on the cathode is precisely the process, as first explained by Shin et al\(^2\), that leads to the desirable micro-porous structure. The fundamentals of the electrochemical deposition process are illustrated in Figure 18.

A polished copper cylinder was used as the cathode and a copper plate was used as the anode. The two electrode surfaces were fixed parallel in the electrolyte at a 20 mm distance. The electrolyte was a solution of 1.5 M sulphuric acid (H\(_2\)SO\(_4\)) and various concentrations of copper sulphate (CuSO\(_4\)). During the deposition a constant DC current was applied, using a precision DC power supply (Thurlby-Thandar TSX3510). The deposition was performed in a stationary electrolyte solution without stirring or N\(_2\) bubbling.

During the deposition, the growth of the dendritic copper structure was blocked by the hydrogen bubbles, wherefore the hydrogen bubbles functioned as a dynamic masking template during the deposition. The hydrogen bubbles depart from the surface, rise and merge into larger bubbles, and as a result the pore size of the deposited copper structure increase with distance from the surface, which can be clearly seen from SEM images of structures fabricated with various deposition time.

The deposition process can be described as a competition between hydrogen evolution and coalescence away from the surface and metal deposition on to the surface. At low current density (<2 A/cm\(^2\)) the frequency and nucleation density of the hydrogen bubbles were low, result-

![Figure 18. Overview of the electrochemical deposition process.](image-url)
ing in a dense dendritic structure without any pores, but where only traces of the hydrogen bubble template could be seen at the SEM images. At increasing current density (≥3 A/cm²), the bubble population, frequency and coalescence increased to such an extent that the bubbles created permanent voids above the cathode and thereby functioned as a masking template, producing the desired structure.

SEM and TEM images of the micro-porous structure and the dendritic sub-structure are shown in Figure 19. Detailed analysis of the dendritic branches showed that the branches comprise nano-sized grains between 10-20 nm.

Figure 19. a-c) SEM images of the micro-porous dendritic structures at various magnifications. d) TEM image of part of dendritic branch.
3.2 Important Fabrication Variables

The features and properties of the micro-porous surface structure with dendritically ordered nano-particles can be controlled and altered, both on a macro, micro and sub-micron level, by employing various fabrication techniques and varying certain manufacturing parameters. The aim is to produce an optimized structure that results in the best heat transfer performance for a specific application.

3.2.1 Annealing

Since the structural stability of the micro-porous surface structure is relatively weak. The usefulness of the structure in real heat transfer applications will therefore be limited. For this reason, the structure was sub-

![Figure 20. Dendritic structure before (left side) and after (right side) annealing. x200 (top), x1000 (middle), and x5000 (bottom) magnification.](image)
jected to an annealing process. Annealing is a heat treatment process which takes place below the melting temperature of the copper and under a reducing atmosphere (hydrogen) to avoid oxidization of the copper. After the annealing treatment, the micro-porous structure remained intact (pore size, thickness of the structure, pore density), but the sub-micron related features of the structure changed, due to the growth of the grain size of the nano-dendritic branch-like features, which can be visually verified in Figure 20. As the grains grew during annealing treatment, the interconnectivity of the grains was also improved, and the grain boundary effect of nanoparticles was reduced. The annealing treatment thus resulted in:

- Improved mechanical stability of the structure, which has been verified in the structural stability tests see 3.3.
- Improved electrical-, and therefore also thermal-, conductivity of the structure.
- Improved boiling heat transfer performance compared to a non-annealed structure.

The determining parameters of the annealing treatment are time and temperature. If the temperature of the annealing treatment is too high, about 650 °C and higher, the structure will melt, as illustrated in Figure 21. Short time and low temperature will have a small effect on the grain growth while higher temperatures for longer times will result in large grain growth, see Figure 22.

Since the annealing treatment changes the nano-scale morphology of the structure, it may be used to produce an optimized structure in terms of the size of deposited features that results in the best heat transfer performance for a specific application. The annealing treatment and its pos-

Figure 21. Micro-porous structure before (left side) and after (right side) annealing at 700 °C for 5 h.
sibility of tailoring the structure makes the structural features this novel enhancement completely different from prior art.

3.2.2 Time and concentration
Three important manufacturing parameters during the electrodeposition process are deposition time, current density and molar concentrations of sulphuric acid and copper sulphate. In summary, longer deposition time and higher molar concentration (up to 0.4 M) result in a thicker structure.

![X 5000 magnification](image1)
![X 10 000 magnification](image2)

300 °C @ 1 h (Se0511301a)

300 °C @ 1 h (Se0511301a)

500 °C @ 5 h (Se0512081a)

500 °C @ 5 h (Se0512081a)

600 °C @ 10 h (Se0512084a)

600 °C @ 10 h (Se0512084a)

Figure 22. Size variation of grain after annealing treatment at various temperatures.
with larger pores. The pore size is measured at the top of the structure, which are the pores seen from SEM images. As illustrated in Figure 23, thickness, pore size and deposition amount are almost linear functions of the deposition time.

3.2.3 Surface Orientation and Geometry

The orientation of the cathode was also affecting the dynamics of the hydrogen evolution and the Cu deposition. All pool boiling results and discussions in this thesis report have been based on an electrochemical deposition process where the cathode is in a horizontal position facing up (0°), but it has been found that the deposition can also take place with the cathode in any position. With the cathode at a vertical angle (90°) the result was almost identical to that of 0°. But, with the cathode facing down (180°) the hydrogen bubbles would not easily escape, but rather coalescence and eventually form a large bubble covering the whole surface. Hence, the Cu deposition was completely obstructed after approximately 25 sec. of deposition. The structure was similar to the ones made at 0° and 90°, but since the growth only continued for 25 sec. there was a limit to the thickness of the structure. Furthermore, since the bubbles coalesced and stayed on the surface at 180° deposition, less current density was needed to create the porous structure.

Figure 23. Various characteristics of the micro-porous structure as a function of time at 1.5M H₂SO₄ and 0.4M CuSO₄.

3.2.3 Surface Orientation and Geometry

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Since the structure may be fabricated on a surface of any direction, it is possible to apply the micro-porous enhancement structure to many different geometries that might be interesting heat transfer applications, such as plate heat exchangers, inside and outside of tubes, fins, etc.

3.2.4 Additives

The pore sizes and wall structures are tunable by using various additives in the electrolyte. One such additive, among many others, is hydrochloric acid (HCl), which had the effect of considerably reducing the elementary branches in the structure and changing the shape of the pores, see Figure 24. In boiling test, the structure fabricated with 50 mM HCl in the electrolyte solution did not perform as well as the surfaces fabricated under same conditions, but without any additives.

3.2.5 Temperature of Electrolyte

By increasing the temperature of the electrolyte to 65 °C from room temperature (20 °C), the pore size of the structure was increased considerably from 47 to 125 µm (3 A/cm², 20 s deposition time, 1.77 cm² substrate area, and 0.4 M H₂SO₄ concentration), see Figure 25. The dendritically formed grains were similar in size. The deposited amount was
also found to increase from 14 to 17 mg. The boiling heat transfer performance of structures fabricated at high electrolyte temperature has not yet been investigated.

3.2.6 Pressure of Electrolyte

By enclosing the electrodeposition set-up in a small stainless steel pressure chamber, it was possible to fabricate the structure under varying pressures. At low pressure, 0.05 bar (abs), the pore size was reduced from 30 µm (at 1 bar) to 22 µm, while the deposition amount increased from 7 mg (at 1 bar) to 11 mg (3 A/cm², 10 s deposition time, 1.77 cm² substrate area, and 0.4 M H₂SO₄ concentration), see Figure 26. The reduction in pore size under lower pressure was unexpected, considering that gas bubbles expand as under lower pressure. It was also observed that the micro-porous structure at low pressures was more homogeneous than at higher pressures, which also can be seen in Figure 26.

At higher pressure, 3.0 bar (abs), the change in pore size was negligible compared to deposition at 1 bar. The deposition amount decrease from 7 mg (at 1 bar) to 4 mg (same deposition conditions as above).
3.3 Mechanical Stability of Structure

An experimental setup has been designed with the purpose to test the mechanical stability of the nano-dendritic micro-porous structure, see Figure 27. Pressurized air was blown over the surface at controlled distance, pressure, angle, and time, see Table 1. The tested structures were fabricated on the top of copper cylinders (also called substrate), with the same dimensions as the cylinders used in the pool boiling tests. The cylinders were weighed before and after fabrication of the structure and af-
After the tests to determine how much of the surface structure that eroded during the tests. Four surface structures which had undergone various treatments were tested and compared. The electrodeposition conditions, during the fabrication of the micro-porous structure, were the same for all four structures (30 s, 0.4M, 20 °C, 3 A/cm²). Tested structures were:

- A non-annealed structure.
- An annealed structure.
- An annealed structure where the substrate had been electroplated (EP) at low current density for 2 minutes before the micro-porous structure was fabricated on the substrate.
- An annealed structure where the substrate had undergone a 2 min etching treatment before the micro-porous structure was fabricated on the substrate.

The results from four tests are shown in Figure 28, where it can be seen that the micro-porous structure which had not undergone annealing was almost completely eroded after the 60 mm distance test, while the annealed structure was least susceptible to erosion throughout the complete range of testing conditions. The electroplating and etching treatments were intended to improve the adhesion between the substrate and the structure. But, since none of the four structures were completely eroded during the tests, the possibly improved adhesion of the EP and

<table>
<thead>
<tr>
<th>Distance between nozzle and surface</th>
<th>5 - 60 mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pressure</td>
<td>7-7.5 bar (rel.), fluctuating over a 1 min period</td>
</tr>
<tr>
<td>Time</td>
<td>180s + 15s startup time</td>
</tr>
<tr>
<td>Surface orientation</td>
<td>Vertical</td>
</tr>
</tbody>
</table>

Figure 28. Results from mechanical stability tests. Intact structure is defined as the weight of structure after test / weight of structure before test.
etched treated structures was not quantified. In other words, the measured decrease in weight of the structures after each test was due to erosion of the top layers of the structures. This is further illustrated in Figure 29 where it can be seen that 94% of the non-annealed structure was eroded already after the 60 mm distance test, but the adhesion of the bottom layer of the structure to the substrate is still intact.

The variation between the annealed structure and the two structures that had undergone electroplating and etching are probably within the error margin of the test and structure fabrication repeatability. It is not likely that these pre-treatments of the substrate would have a negative effect on the mechanical stability of the fabricated structure.

3.4 Electrical and Thermal Conductivity of Structure

As presented in section 4.4, the improved boiling performance of the annealed structures is thought to be due to the improved thermal conductivity of the annealed structures. The improved thermal conductivity of the structures is a result of the grain growth and increase connectivity of the grains that takes place during the annealing process. To confirm this theory, the thermal conductivity, before and after annealing of the structure, needed to be measured, but it is rather complicated to measure the thermal conductivity of a thin film, and especially of a thin porous structure such as the nano-dendritic micro-porous structure. Therefore, an indirect method was developed where the electrical conductivity of the structure was measured, before and after annealing.
Since the electrical conductivity is proportional to the thermal conductivity of a metal, according to the Wiedemann-Franz law, this method ought to give an indication of how the thermal conductivity of the structure, $\lambda$, is changed after the annealing treatment. Since the electrical conductivity is inversely proportional to the electrical resistance, $R$, the relationship is:

$$\frac{\lambda_{\text{annealed}}}{\lambda_{\text{non-annealed}}} = \frac{R_{\text{non-annealed}}}{R_{\text{annealed}}}$$  \hspace{1cm} (1)

To be able to measure the electrical resistance of the micro-porous structure, the structure was fabricated in-between two thin copper plates, which had been electrically insulated and fixated with a ceramic insulation paste, see Figure 30. One plate functioned as anode and one as cathode and during the electrodeposition step, the structure grew until it connected to the cathode, resulting in short circuiting of the two plates.

The electrical resistance between the two plates was found to decrease approximately 20 times (from 0.23 to 0.01 $\Omega$) after annealing (500 °C for 5 h). Thus, a strong indicative for a better thermal conductivity of the annealed structure has been presented. One large uncertainty of this test is if there exists a relatively large electrical connection resistance between the structure and the anode. If this is the case, it is possible that the annealing treatment substantially lowers this electrical connection resistance and hence the reduction in total electrical resistance would not be due to a reduction of the resistance through the structure.
4 Pool Boiling Experiments

4.1 Experimental Apparatus and Procedure

4.1.1 General set-up
The central part of the experimental pool boiling set-up is illustrated in Figure 31. The boiling tests were conducted inside a boiling chamber, insulated with 10 mm thick foam insulation, and with an internal volume of 50 dm$^3$. The widely used hydrofluorocarbon refrigerant: R-134a, was employed as test liquid in the experiment. Vapor from the boiling chamber was condensed in an insulated condensing chamber, located above the boiling chamber. The condenser was cooled by a secondary loop. The refrigerant in the secondary loop was cooled by an external compressor driven refrigeration cycle. Saturated liquid from the condenser flowed back into the boiling chamber by the influence of gravity.

A pressure transducer (Druck, PDCR911), measuring the vapor pressure of the boiling chamber, was connected to a logger (Cambell, M 21x), which every second regulated the condenser effect by opening and closing a magnetic valve on the secondary refrigerant loop, to maintain constant pressure in the boiling chamber. With this set-up, the desired pressure level could be maintained within a range of ±2 mbar during the test.

Figure 31. Schematic overview of test apparatus, test objects and teflon insulation block.
Two thermocouples (Pentronics, HF/D-30, type T) were employed to measure the liquid temperature and were placed in the middle of the bulk test liquid, away from the bubble pathway.

4.1.2 Test Object
The nanostructured micro-porous surface was fabricated, before assembly of test object, on one end of a 6 mm thick copper cylinder with a diameter of 15 mm. High resistance heating wire (Degussa, Isoliert Thermofraht) 14.8 $\Omega$/m was used to make a coil heater, which was attached to the base of the copper cylinder using thermoconducting silver epoxy (Circuitworks, CW2400) as illustrated in Fig. 2. The electrical resistance of the heater was measured with a bench multimeter (Fluke 45 dual display) to about 3.50 $\Omega$. The resistance of the heater has been tested at different heat fluxes within the range of this study and found to be constant.

The copper cylinder was fitted in a cylindrical Teflon casing with a slot machined for the copper cylinder. To prevent edge nucleation around the perimeter of the surface, the opening of the slot was 14 mm in diameter, creating a 0.5 mm long edge overlapping the copper surface. Epoxy adhesive was carefully applied on the inside of the Teflon casing edge to ensure a flush-mounted copper cylinder. One 1 mm diameter hole for a $\varnothing$0.8 mm thermocouple was drilled into the center of the copper cylinder, 2 mm below the surface. The hole was filled with thermoco nductive paste (Electrolube, HTC Plus) before insertion of thermocouple and then sealed with fast curing epoxy to prevent test liquid

![Figure 32. Test object design including a cross-sectional illustration of the heat dissipation in the test object holder.](image)
from entering the thermocouple well. The void space in the Teflon casing around the heater was filled with epoxy to create a test object with a plane bottom.

Each test object was tightly fitted in one of the machined slots in an insulating Teflon block (λ_{teflon}=0.35 Wm⁻¹K⁻¹) with the dimensions 240 mm x 40 mm x 85 mm. Up to three test objects could be run simultaneously. Three ∅1.0 mm holes, for each test object, were drilled through the Teflon block to make room for the two power leads and the thermocouple. The reference surface was a copper surface polished with 240 grade emery paper, to simulate a plain machined surface.

4.1.3 Test Procedure
The Teflon block, with the test objects, was placed horizontally at the bottom of the boiling chamber. A vacuum pump was used to evacuate the chamber to a pressure of less than 8 mbar, before it was filled with the test liquid.

Before commencing the test, the pressure (and the corresponding temperature of the test liquid) was lowered to 0.5 bar less than desired pressure level. This was done in order to reduce the vapor entrapped in the surface structure and thereby gave all tests the same pressure history, which was important for the repeatability of the results, as confirmed by Gallagher and Winterton. After the desired pressure level had been attained, a waiting period of about 2 hours was needed before the temperature difference between the test liquid and the test objects was less than 0.1 °C. The heaters of the test objects, connected in series, were supplied with DC power from a computer controlled power supply (Instek, PSP-405). The current was measured with the power supply unit. The logger and the power supply were controlled with computer software (HPVee, 5.0) with values recorded every 3 seconds.

The test series were initiated at a heat flux of 5 W/cm², upon which the whole surface rapidly erupted into nucleate boiling. Heat flux was then increased to a maximum of 11 W/cm² from which it was lowered in steps of approximately 1 W/cm² per measurement point. Presented values are taken from decreasing heat flux. Temperatures were sampled every third second and when 20 consecutive readings (60 s) from each thermocouple were within an interval of 0.1 °C, the average values of the readings were recorded and the heat flux was changed to the next measurement point. All test runs were repeated two times with repeatability within the uncertainty limits.
4.2 Uncertainty Estimates

4.2.1 Methodology

The methodology of Kline and McClintock (1953)\(^4\) to describe uncertainty in experiments is widely spread and accepted, e.g. by American Society of Mechanical Engineers, and has therefore been used in the following uncertainty analysis. The primary objective, of the analysis of experimental data, has been to assess the uncertainty of the heat transfer coefficient. The heat transfer coefficient is a function of five independent variables, which are assumed to be normally distributed and with uncertainty intervals presented in Table 2.

The relation between the uncertainty interval for variable, \(wx_i\), and the uncertainty for the heat transfer measurement result, \(wh\), is calculated by means of the combination of variances:

\[
w_h = \sqrt{\sum_{i=1}^{m} \left( \frac{\partial h}{\partial x_i} \cdot wx_i \right)^2}
\]

Where the function for the heat transfer coefficient, \(h\), was defined as:

\[
h = \frac{\dot{Q}}{A \cdot \Delta T_{\text{surface}}} = \frac{R \cdot I^2}{\pi \cdot d^2 \cdot \Delta T - R \cdot I^2 \cdot \frac{\Delta x}{\lambda_c}}
\]

Table 2. Variables and their uncertainty estimates.

<table>
<thead>
<tr>
<th>Variable, (x_i)</th>
<th>Uncertainty interval for variable (20:1 odds), (wx_i)</th>
<th>Source of uncertainty estimate</th>
</tr>
</thead>
<tbody>
<tr>
<td>Current ((I))</td>
<td>±0.1% x I</td>
<td>User manual to Instek PSP-405</td>
</tr>
<tr>
<td>Resistance ((R))</td>
<td>±0.05% x R</td>
<td>Data sheet for Fluke 45</td>
</tr>
<tr>
<td>Diameter of surface ((d))</td>
<td>≤0.1 mm</td>
<td>Calibration against precision micrometer</td>
</tr>
<tr>
<td>Temperature difference ((\Delta T))</td>
<td>≤0.1 ºC</td>
<td>Calibration and experience, see section 4.3.3.</td>
</tr>
<tr>
<td>Location of thermocouple ((\Delta x))</td>
<td>≤ 0.1 mm</td>
<td>Calibration against precision micrometer</td>
</tr>
</tbody>
</table>
The temperature difference, between the boiling surface and the bulk liquid, $\Delta T_{surface}$ had to be calculated, using Fourier’s law of conduction:

$$\Delta T_{surface} = \Delta T - \Delta T_{corr} = \Delta T - \frac{Q \cdot \Delta x}{A \cdot \lambda_c}$$  

(4)

Where $\Delta T$ is the temperature difference between the measurement point, at a distance of approx. 2 mm under the boiling surface (see Figure 32), and the average bulk liquid temperature. $\Delta T_{corr}$ is the calculated temperature drop between the boiling surface and the location of the thermocouple.

After partial derivation of equation 3, variance contribution of each variable was calculated according to:

$$\frac{\partial h}{\partial I} w_I = \frac{R \cdot I^2 \cdot \pi \cdot d^2 \cdot \Delta T}{\left( \frac{\pi \cdot d^2}{4} \cdot \Delta T - R \cdot I^2 \cdot \frac{\Delta x}{\lambda_c} \right)^2} \cdot w_I$$  

(5)

$$\frac{\partial h}{\partial R} w_R = \frac{I^2 \cdot \Delta T \cdot \frac{\pi \cdot d^2}{4}}{\left( \frac{\pi \cdot d^2}{4} \cdot \Delta T - R \cdot I^2 \cdot \frac{\Delta x}{\lambda_c} \right)^2} \cdot w_R$$  

(6)

$$\frac{\partial h}{\partial d} w_d = \frac{-R \cdot I^2 \cdot \pi \cdot d \cdot \Delta T}{\left( \frac{\pi \cdot d^2}{4} \cdot \Delta T - R \cdot I^2 \cdot \frac{\Delta x}{\lambda_c} \right)^2} \cdot w_d$$  

(7)

$$\frac{\partial h}{\partial \Delta T} w_{\Delta T} = \frac{-R \cdot I^2 \cdot \frac{\pi \cdot d^2}{4}}{\left( \frac{\pi \cdot d^2}{4} \cdot \Delta T - R \cdot I^2 \cdot \frac{\Delta x}{\lambda_c} \right)^2} \cdot w_{\Delta T}$$  

(8)
\[
\frac{\partial h}{\partial \Delta x} \cdot W_{\Delta x} = \frac{-R^2 \cdot I^4}{\lambda_c} \left( \frac{\pi \cdot d^2}{4} \cdot \Delta T - R \cdot I \cdot \frac{\Delta x}{\lambda_c} \right) \cdot W_{\Delta x}
\]  

(9)

4.2.2 Results and Analysis

Table 3 presents the uncertainty contribution to the heat transfer coefficient from each of the variables for two different surfaces; one reference surface and one high performance enhanced surface.

To conclude, the uncertainty of the measurements of the heat transfer coefficient is highly dependent on the measurement of the temperature difference, \(\Delta T\), which in turn will be a function of heat flux and boiling performance of the surface. Higher heat flux results in higher temperature differences, which is followed by better accuracy, whereas better performing surfaces results in lower temperature differences which in turn results in worse accuracy.

For enhanced surfaces at high heat flux, >5 W/cm², the heat transfer coefficient can usually be stated with an accuracy better than ±10% whereas at low heat fluxes <1 W/cm², the accuracy decreases substantially for high performance surfaces and can in extreme cases (lowest heat flux, best performing surface) be over ±80% since the measured temperature difference is approaching 0.1°C.

Table 3. Uncertainty contribution to the heat transfer coefficient from different variables

<table>
<thead>
<tr>
<th></th>
<th>Reference Surface</th>
<th>Enhanced Surface</th>
</tr>
</thead>
<tbody>
<tr>
<td>(q) [W/cm²]</td>
<td>1.0</td>
<td>10.0</td>
</tr>
<tr>
<td>(h) [W cm⁻² K⁻¹]</td>
<td>0.2</td>
<td>1.0</td>
</tr>
<tr>
<td>Current ((I))</td>
<td>±0.2%</td>
<td>±0.2%</td>
</tr>
<tr>
<td>Resistance ((R))</td>
<td>±0.05%</td>
<td>±0.05%</td>
</tr>
<tr>
<td>Diameter of surface ((d))</td>
<td>±1.4%</td>
<td>±1.4%</td>
</tr>
<tr>
<td>Temperature difference ((\Delta T))</td>
<td>±2.2%</td>
<td>±1.0%</td>
</tr>
<tr>
<td>Location of thermocouple ((\Delta x))</td>
<td>±0.06%</td>
<td>±0.3%</td>
</tr>
<tr>
<td>Combined uncertainty ((w_h/r)):</td>
<td>±2.7%</td>
<td>±1.8%</td>
</tr>
</tbody>
</table>

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4.2.3 Uncertainty in Temperature Measurements

As seen in the preceding analysis, the uncertainty in the temperature measurements gives the largest contribution to the overall uncertainty of the heat transfer coefficient. Therefore, a closer look at the estimated uncertainty may here be warranted.

According to the manufacturer (Campbell Scientific), the resolution for voltage measurement in the data logger corresponded to 0.008 °C and the function error (conversion from voltage to temperature) was less than ±0.001 °C in the applicable range. At thermally stable conditions, all temperatures in the experimental set-up were within ±0.04 °C, as illustrated in Figure 33, and the standard deviation for each thermocouple was less than 0.02 °C. Calibrations were regularly run before and after each test run. Since these expected components of uncertainty are considered as stochastic variables, the combined uncertainty of the temperature measurements are calculated by means of combination of variances according to:

\[
\sigma_{\Delta T} = \sqrt{\sigma_{\Delta T,\text{resolution}}^2 + \sigma_{\Delta T,\text{function}}^2 + \sigma_{\Delta T,\text{calibration}}^2 + \sigma_{\Delta T}^2}
\]

(10)

\[
\sigma_{\Delta T} = \pm \sqrt{0.008^2 + 0.001^2 + 0.04^2 + 0.02^2} = \pm 0.045° C
\]

Hence, the uncertainty interval for the temperature difference (\(\Delta T\)) has been conservatively estimated to ±0.1 °C (20:1 odds).

Figure 33. Thermocouples at thermally stable conditions.
4.3 Heat Loss Calculations

4.3.1 Methodology
Heat losses through the teflon insulation block has been calculated using a finite element solver (FEMLAB 3.0) and an engineering equation solver program with refrigerant property data (EES, v7). An iterative procedure has been employed including following steps;

1. (FEMLAB 3.0): Based on input values (geometry, number and position of test objects, heat transfer coefficients and heat input), the average temperature of the boiling copper surfaces (1, 2 or 3) and all the six sides of the teflon insulation block where calculated.

2. (EES, v7): The temperatures, from step 1, were used to calculate the heat transfer coefficients for each side of the teflon insulation using various free convection correlations, see below.

3. The heat transfer coefficients, from step 2, served as the new input for step 1 and the iteration was performed until temperatures converged to their final values.

4. (FEMLAB 3.0): Finally, the heat flux through the boiling surfaces were compared to the losses through the sides of the teflon insulation block.

Following correlations were used to calculate the heat losses through the six sides of the teflon insulation:

**Vertical surfaces**, LeFevre (1956)\(^5\):

\[
\overline{Nu_L} = \frac{\bar{h} \cdot L}{k} = \frac{4}{3} \left( \frac{Gr_L}{4} \right)^{\frac{1}{4}} \cdot g(Pr) \tag{11}
\]

Where the Grashofs number, \(Gr_L\), is defined as:

\[
Gr_L = \frac{g \cdot \beta \cdot (T_s - T_s) \cdot L^3}{\nu^2} \tag{12}
\]

and the dependence on the Prandtl number, \(Pr\), is given by the function:
\[ g(Pr) = \frac{0.75 \cdot Pr^{\frac{1}{4}}}{\left(0.609 + 1.221 \cdot Pr^{\frac{1}{2}} + 1.238 \cdot Pr\right)^{\frac{3}{4}}} \]  \hspace{1cm} (13)

**Horizontal surface**, Lloyd and Moran (1973)\(^6\) and Goldstein et al. (1974)\(^7\):

- **Facing up**: \[ \overline{Nu}_L = 0.15 Ra_L^{\frac{1}{3}} \left(10^7 \leq Ra_L \leq 10^{11}\right) \]  \hspace{1cm} (14)

- **Facing down**: \[ \overline{Nu}_L = 0.27 Ra_L^{\frac{1}{4}} \left(10^5 \leq Ra_L \leq 10^{10}\right) \]  \hspace{1cm} (15)

Where the Rayleigh number, \(Ra\), is defined as:

\[ Ra_L = Gr_L \cdot Pr \]  \hspace{1cm} (16)

and the characteristic length is defined as:

\[ L = \frac{A_s}{P} \]  \hspace{1cm} (17)

Where \(A_s\) is the plate surface area and \(P\) is the perimeter.
4.3.2 Results and Analysis

The relative heat loss for two selected experimental test runs is presented in Table 4 and visualizations of the temperatures in the test object and the insulation are illustrated in Figure 34 and Figure 35.

As illustrated in Table 4, the relative heat loss through the teflon insulation is a negative function of heat input. This can be explained by that the heat transfer coefficient of the surfaces are not increasing as quickly as the heat input, hence a larger fraction of the heat input will be lost through the insulation. As expected, the heat loss from tests with the enhanced surfaces was significantly lower than that from the tests with the reference surface. The higher heat transfer coefficients of the enhanced surfaces result in lower temperature of the test object which is followed by smaller heat losses through the insulation. This can be clearly seen in comparing Figure 34 and Figure 35.

![Figure 34](image1.png)
![Figure 35](image2.png)

*Figure 34. (FEMLAB) Visualizations of temperatures in the reference surface position 1 and in the insulation block @ 10 W/cm².*
The heat transfer coefficients presented in this work, have not been adjusted for the heat loss through the insulation, and are therefore slightly overstated.

4.4 Experimental Results

Table 5 presents a summary of some structure characteristics of the seven surfaces that have been tested. Figure 37 shows boiling curves of the eight different surfaces, including the reference surface. To further illustrate the boiling characteristics of the different structures, Figure 36 has been included, which shows the heat transfer coefficient vs. heat flux. As seen in Figure 37, the reference surface closely follows the well-known correlation suggested by Cooper8 (4 bar, 2 R). All of the enhanced surfaces sustained nucleate boiling at lower surface superheat than the reference surface. The 120µm-annealed (120µm-a) and 220µm-a surfaces performed better than their non-annealed counterparts up to 7 W/cm², above which the non-annealed surfaces performed slightly

<table>
<thead>
<tr>
<th>Surface id.</th>
<th>80</th>
<th>120</th>
<th>220</th>
<th>80a</th>
<th>120a</th>
<th>220a</th>
<th>265a</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thickness [µm]</td>
<td>80</td>
<td>120</td>
<td>220</td>
<td>80</td>
<td>120</td>
<td>220</td>
<td>265</td>
</tr>
<tr>
<td>Pore Diameter [µm]</td>
<td>30</td>
<td>45</td>
<td>65</td>
<td>30</td>
<td>45</td>
<td>65</td>
<td>105</td>
</tr>
<tr>
<td>Pore Density [N/mm²]</td>
<td>470</td>
<td>150</td>
<td>100</td>
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<td>Annealing</td>
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<td>No</td>
<td>No</td>
<td>5h (@500°C)</td>
<td>5h (@500°C)</td>
<td>5h (@500°C)</td>
<td>5h (@500°C)</td>
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Figure 35. (FEMLAB) Visualizations of the temperatures in two enhanced surfaces in positions 1 and 2 and in the insulation block @10 W/cm².

Table 5. Tested surfaces.
better. At low heat flux, the annealed surfaces, 120µm-a and 220µm-a, performed exceptionally well with surface superheats of approx. 0.3 °C at 1 W/cm². This is to be compared to 4.4 °C for the reference surface at the same heat flux, which is an improvement of the HTC with over 16 times. At high heat flux, 10 W/cm², non-annealed surface, 120µm, had a superheat of 1.4 °C, when the reference surface was recorded at 9.4 °C, an improvement of almost 7 times of the HTC.

![Figure 36. Heat transfer coefficient vs. heat flux.](image)

![Figure 37. Boiling curves of enhanced surfaces and reference surface.](image)
4.4.1 Analysis

The remarkably effective heat transfer capabilities of the structure are suggested to be caused by the following three characteristics of the structure; suitable vapor escape channels, high porosity, and dendritic branch formation.

Suitable vapor escape channels. The pores in the structure, seen from a top view in Figure 19 and in Figure 20, are believed to act as vapor escape channels during the boiling process. Since the pores are formed by the template of the rising hydrogen bubbles during the electrodeposition process, Figure 18, trails of growing and interconnected pores are left, shaping channels which penetrate the whole structure from the base to the top. This feature, together with the high pore density: 470, 150, and 100 per mm$^2$ at different heights of the structure: 80, 120, and 220$\mu$m respectively, ensure that the vapor produced, during evaporation inside the structure, can quickly be released with low resistance from the dendritic structure. The interesting resemblance between the manufacturing process of the structure and the boiling phenomena itself is striking. The departing hydrogen bubbles are seeking the lowest resistance path, thus creating low impedance vapor escape channels.

SEM images of the 80$\mu$m-a structure, reveal that the increase of dendritic grain size has defected the shape of many of the small surface pores in the structure, thereby adding resistance to vapor and liquid flow as compared to the non-annealed structure of same thickness. Hence the 80$\mu$m-a surface performed worse than its non-annealed counterpart. This observation of the 80$\mu$m-a structure confirms that the vapor escape channels are important for effective mass transport.

High porosity. The unusually high porosity of the structure (calculated by comparing the measured density of the structure with the density of copper) promotes the influx of liquid and the outflow of vapor. With low hydraulic resistance, one-phase liquid flow within the structure could also contribute considerably to the transfer of sensible heat. Laminar flow through small hydraulic diameters is associated with large heat transfer coefficients and is likely to occur within the highly porous structure and may also be enhanced by the pumping action from active nucleation sites. Since the sensible heat transfer can account for a significant part of the heat transferred from enhanced structures, as shown for a porous particle layer by Kim et al. and for the Thermoexcel-E surface by Nakayama et al., the high porosity of the structure is a central feature which contributes to its efficient boiling performance.
Figure 38 shows the result of an almost 90 hour long boiling test at 5 W/cm². The stability of the superheat of the surface, only an increase of 0.05 °C was recorded, which was reversed upon restart, indicates that no major patches of vapor are formed inside the structure, but that the liquid supply through the porous structure is efficient (otherwise vapor patches would grow in size and create local dry spots on the surface). The long time boiling test also shows the durability of the structure.

**Dendritic branch formation.** The structure, as seen in the high magnification images in Figure 19 and in Figure 20, features an exceptionally large surface area, which could facilitate large formations of thin liquid films with high evaporation rates, as initially suggested by O’Neill et al.¹¹ for the porous High-Flux surface and visually documented by Nakayama et al.¹⁰. Furthermore, the dendritic branch formations in the structure, with its jagged cross-section, may generate a long three-phase-line formed by intersection of the vapor-liquid interface with the dendritic branches, which Mitrovic¹²,¹³ presented as an important boiling enhancing mechanism in protruding micro-structures, see Figure 6.

The boiling characteristics of the annealed vs. the non-annealed surfaces seem to indicate that there was an influence of the surface irregularities on the dendritic branches, formed by the micron to sub-micron scale particles. The larger surface area of dendritic branches of the non-annealed structures, as seen in Figure 20, could be the explanation to the continued increase of the HTC, even at higher heat flux, as seen in Figure 36.

At lower heat flux, the improved interconnectivity of the grains, on a nano- and micro scale, resulting in increased thermal conductivity of the annealed structures, is suggested as an explanation to the improved per-
formance of the annealed structures over the non-annealed structures. Among the annealed surfaces, thicker structures performed better than thinner ones, but for the non-annealed structures, the performance was diminishing with structures of greater thickness than 120 µm. This behavior could be related to the thickness of the superheated thermal boundary layer, as described by Chang and You\textsuperscript{14}. Additional height of the structure, beyond the thickness of the thermal boundary layer, increases the hydraulic resistance to the vapor and liquid flow inside the structure and therefore inhibits the heat transfer performance of the structure. The thickness of the superheated thermal boundary layer is a function of the thermal conductivity of the structure. Hence, the annealed structures, with their improved thermal conductivity, displayed better performance with increased thickness, even beyond 120 µm.
4.5 References


5 Next Steps

So, we have seen that the novel micro-porous surface structure comprising dendritically ordered nano-particles of copper performs exceptionally well in pool boiling experiments, but now two important questions arise: How well does this novel structure perform in real applications, and particularly in flow boiling applications? What is it that makes this novel micro-porous structure so suitable for pool boiling?

In an attempt to answer these two queries, a number of experiments have been planned:

- **Thermal tests of plate heat exchanger** (project already on-going). An attempt will be made to up-scale the fabrication process and apply the novel micro-porous structure to the boiling side of a regular plate heat exchanger. The heat exchanger will then be experimentally tested with R134a as refrigerant under various heat and mass fluxes. This experiment will give a good indication of the usefulness of the structure in large heat pumping systems.

- **Flow boiling in enhanced micro-channels** (project already on-going). Two-phase flow and heat transfer in micro-channels is a research field which currently receives much attentions and the possible applications of such small heat exchangers are numerous. The novel micro-porous structure will be applied to the inside of a 400 µm high and 60 mm wide rectangular copper channel, followed by flow boiling tests of the one-channel heat exchanger where the combination of micro-channel and enhanced surface structures will be investigated.

- **Evaporator in Thermosyphon.** The heat fluxes from electronic components are steadily increasing and have now, in some applications, reached levels where air cooling is no longer sufficient. Hence, new methods of cooling of electronics are necessary, of which one is to use a closed loop two-phase thermosyphon system. The temperature difference between the evaporator wall and the refrigerant is most important in the thermosyphon system. This is one
application where the novel micro-porous surface might find itself very useful. The tests will be conducted in a regular PC equipped with a thermosyphon loop which cools the processor. Two different refrigerants, R123 and Isobutane, will be tested.

- **Enhancements of Critical Heat Flux in Pool Boiling** (project already on-going). Critical heat flux is an important parameter in designing cooling systems for certain applications, not the least in the cooling of electronics. At very high heat fluxes, a regular boiling surface will transition into the film boiling regime (the so called critical heat flux, CHF) and thereby experience a rapid temperature increase of several hundred degrees. This would result in a breakdown of the electronic component, but with enhanced boiling techniques, the CHF can be increased and thus ensure continuous cooling of the component. The tests will be run in R134a and in FC-72 (see below).

- **High speed Pool Boiling Visualization in FC-72.** FC-72 is a dielectric refrigerant which is used in electronics cooling applications where the hot component can be in direct contact with the cooling liquid without risk of short circuiting the unit. One advantage of using FC-72 is that there is much data available from enhanced boiling tests with FC-72, so boiling performance tests with the novel micro-porous surface can be readily compared with other enhancement structures. To get a better understanding of the boiling enhancement mechanisms involved with the novel structure, the experiments will be filmed and analyzed, using a high speed camera.
### 6 Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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<tbody>
<tr>
<td>$A$</td>
<td>Area</td>
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<tr>
<td>$d$</td>
<td>Diameter</td>
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<tr>
<td>$\Delta T$</td>
<td>Temperature difference</td>
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<tr>
<td>$\Delta x$</td>
<td>Distance between of thermocouple and surface</td>
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<tr>
<td>$Gr$</td>
<td>Grashofs number</td>
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<tr>
<td>$h$, HTC</td>
<td>Heat transfer coefficient</td>
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<td>Current</td>
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<tr>
<td>$L$</td>
<td>Length</td>
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<tr>
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<td>Molar concentration (moles solute/liter solution)</td>
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