Investigation of Separate Effects of Surface Condition on Subcooled Flow Boiling Heat Transfer

by

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Abstract

In Pressurized Water Reactor (PWRs), heat generated by nuclear fissions is effectively transferred to the water coolant by subcooled flow boiling. The maximum reactor power is limited by the critical heat flux (CHF), at which the boiling crisis occurs. Understanding the mechanisms that trigger this boiling crisis and predicting the CHF limit is key to the safety and efficiency of nuclear reactors. The CHF limit depends on cladding material, thickness, and surface conditions. Importantly, the surface of a fuel rod cladding evolves during operation due to oxidation and crud deposition. Many studies have investigated the effects of surface properties (e.g., surface roughness, wettability, and porosity) on boiling heat transfer and CHF. Still, the results of these investigations are not always in agreement with each other. We believe that the reason for these discrepancies is due to the lack of control of the surface conditions.

This thesis aimed at developing experimental capabilities and protocols to conduct “separate effect” studies and investigating the real effect of surface oxidation and Accident Tolerant Fuel (ATF) coatings on subcooled flow boiling heat transfer. To that end, we prepared Zircaloy-4 heaters that mimic the commercial PWRs fuel cladding, and conducted subcooled flow boiling experiments at 1 bar, 10 K subcooling, and 1000 kg/m²s mass flux, using high-resolution high-speed video (HSV) and infrared (IR) diagnostics. A computational model solving a 3-D inverse conduction problem was developed to post-process infrared (IR) measurements. An HSV post-processing approach including a deep-learning tool, U-net, and a global optical flow algorithm was proposed to quantify boiling parameters from HSV images. The parameters were incorporated into a heat flux partitioning model, where we introduced a term to account for the non-symmetric growth of the microlayer.

The experimental results showed that groove pattern, average roughness, and wettability do not affect subcooled flow boiling. Instead, they suggest that the process is determined by the location, size, and shape of cavities, and that micro-scale surface modifications (e.g., porous cracks) or nano-scale structures play a crucial role in the formation of active nucleation cavities and modify the bubble dynamics. A key takeaway from this study is that, to elucidate how surface modifications affect boiling heat transfer, one should carefully examine how the surface morphology changes at both the micro- and nano-scale and how the surface preparation process affect the formation of cavities.

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Title: Assistant Professor of Nuclear Science and Engineering
Thesis Reader: Jacopo Buongiorno, Ph.D.
Title: TEPCO Professor of Nuclear Science and Engineering
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Chapter 1

Introduction

1.1 Motivation and Objectives

In the upper part of a pressurized water reactor (PWR) core, heat generated by nuclear fissions is transferred to water by subcooled flow boiling heat transfer, which occurs on the surface of the fuel rod cladding. The maximum reactor power is limited by the efficiency of this heat transfer process. Higher reactor power induces more vigorous boiling and higher heat transfer rate. However, if the local heat flux exceeds a critical limit, the so called critical heat flux (CHF), bubbles merge to form a large irreversible vapor blanket. This vapor blanket leads to a sudden reduction of the boiling heat transfer efficiency, and a rapid escalation of the cladding temperature, which may result into damage or even melting of the fuel cladding. Therefore, understanding the mechanisms that trigger this boiling crisis and predicting the CHF limit is key to the safety and efficiency of nuclear reactors.

The CHF limit depends on several factors. Primarily, it depends on the reactor operating condition, e.g., pressure, sub-channel geometry, mass flux, inlet subcooling and heat flux distribution. Additionally, it depends on cladding material, thickness, and surface conditions, e.g., morphology and wettability. Importantly, the surface of a fuel rod cladding evolves during operation. Its properties change due to oxidation in high temperature water (~350 °C for PWR), radiation, and dissolved chemicals.

Many studies have investigated the effects of surface properties (e.g., surface roughness, wettability, and porosity) on boiling heat transfer and CHF (e.g., see Refs. [1-11]). Still, the results of these investigations, however interesting, are not always in agreement with each other, either qualitatively or quantitatively. We believe that the reason for these discrepancies is due to the lack of control of the surface conditions. In other words, we suspect that “separate effect” studies may not always “separate the surface effect” accurately.
To support this statement, we show in Figure 1.1 (a) high-speed video (HSV) images at 1 MW/m² and boiling curves that we obtained in the exact same subcooled flow boiling conditions (i.e., operating the same flow loop with DI water at 1 bar, with 10 K of subcooling and a mass flux of 1000 kg/m²·s) using two nominally identical Zircaloy-4 samples (A and B). These boiling surfaces were prepared using the same fabrication procedure (i.e., randomly polishing the samples with 240-grit sandpaper with a force of 20 N, at 200 rpm for 2 minutes) and cleaning protocol (i.e., sonicating the samples in Acetone for 2 minutes, and then rinsing with Ethanol, Isopropanol, and DI water).

![Figure 1.1](image)

**Figure 1.1** Demonstration of different boiling behavior on two nominally identical rough Zircaloy-4 surfaces: (a) High-speed video images at 1 MW/m² (left) and boiling curves (right) and (b) bidimensional images of the surface profile taken with an optical profilometer (left) and associated distributions (right). Note that the top figure (a, right) shows boiling curve for 16 nominally identical samples, including sample A and B.

However, while these two samples have the same wettability (i.e., the contact angle, CA, of both surfaces is ~50°) and average surface roughness (Sa ~0.3 μm), they have different boiling dynamics (see high-speed video images in Figure 1.1 (a)), and different heat transfer coefficient (HTC) and CHF (see Figure 1.1). Sample A has higher HTC and lower CHF with small bubbles and high nucleation site density. Sample B has lower HTC and higher CHF with large bubbles and low nucleation site density. Note that Figure 1.1 (a) shows boiling curves of 16 different
samples, including samples A and B, all prepared with the same fabrication procedure and all having the same wettability and roughness. Clearly, while all these surfaces are nominally identical, their boiling curve is very different. We made similar observation studying the boiling behavior on mirror polished, ultra-smooth surfaces, as shown in Figure 1.2.

![Ultra-smooth surface (S<sub>a</sub> < 40 nm)](image)

**Figure 1.2** Demonstration of different boiling behavior on two nominally identical ultra-smooth Zircaloy-4 surfaces. From left to right: bidimensional image of the surface profile taken with an optical profilometer, high-speed video images of the boiling process at 1.2 MW/m<sup>2</sup> on samples C and D, and boiling curves.

These two examples demonstrate that even nominally identical surfaces can have different boiling heat transfer coefficient and CHF (everything else being the same). Crucially, these differences (in the order of 0.5 MW/m<sup>2</sup>) can be larger than the differences often attributed to surface effects, e.g., oxidation. Also, and importantly, these examples confirm that surface roughness and morphology are not necessarily good indicators of the boiling performance. Instead, they may support the hypothesis that other factors determine the efficiency of the boiling process, e.g., the density of nucleation sites, their location, size, and shape.

However, these observations suggest that a proper “separate effect” study of oxidation effects should be conducted using a special experimental protocol where the same exact sample is used to study the dynamic of the boiling process at different stages of oxidation. Likewise, the effect of other surface modifications, e.g., Cr or other Accident Tolerant Fuel (ATF) coatings, should be characterized by studying the boiling process on the same sample, before and after the coating is applied.
This thesis aimed at developing experimental capabilities and protocols to conduct such “separate effect” studies and investigating the real effect of surface oxidation and ATF coatings on boiling heat transfer.

Importantly, while in the past most studies have focused on pool boiling conditions, we have focused our attention on subcooled flow boiling. To achieve this goal, we have developed image processing techniques to extract boiling parameters (e.g., nucleation site density, bubble growth and wait time, and bubble size) from high-speed video images of the boiling process and, based on these parameters, analytic models to predict the boiling heat transfer coefficient (e.g., using a heat flux partitioning model).

We expect that the data and the scientific understanding achieved in this study will support modeling efforts to improve the prediction of subcooled flow boiling heat transfer and CHF in nuclear reactors.
1.2 Background

1.2.1 Effects of Surface Finish on Boiling Heat Transfer

Many studies have investigated the effects of surface roughness on saturated pool boiling heat transfer coefficient (HTC) and CHF using various materials and working fluids. Berenson [1] claimed that the pool boiling HTC depends on the number of nucleation sites with size appropriate for nucleation, regardless of the surface average roughness. Generally, rougher surfaces may have a larger number of such cavities, so their HTC is commonly thought to be higher. However, even smooth surfaces can have high HTC if they have a large enough number of cavities whose size is within the range favorable for bubble nucleation, suggesting that surface finish or average roughness are not universal indicators of the CHF.

Ramilison, et al. [2] gathered existing pool boiling CHF data and derived an empirical correlation to estimate the pool boiling CHF as function of surface roughness, \( r \), and contact angle, \( \theta \). According to this correlation, the CHF is strongly dependent on surface wettability \( (\propto \theta^3) \), but not as much on roughness \( (\propto r^{0.125}) \). The influence of roughness is important only at low heat fluxes, i.e., near the onset of nucleate boiling.

Pioro et al. [3] reviewed existing pool boiling data to elucidate how boiling surface parameters affect nucleate boiling heat transfer. They came to the same conclusion as in Ref. [1]. Although roughness is the most common parameter used to characterize the surface, it does not, in fact, represent the real surface microstructure. Instead, cavity shape, size, and density are the most important factors to determine bubble nucleation and dynamics of the boiling process. The average roughness can be a good indicator for the HTC only when the roughness change leads to a change of the active cavities density. However, if surface is very rough, the HTC can increase due to rough structures acting as micro-fins.

Jones et al. [4] observed that a roughened surface has a larger number of nucleation site, smaller bubble, higher nucleation frequency, and higher HTC. The HTC increased by increasing the average roughness from 0.0027 \( \mu \)m to 1.08 \( \mu \)m, but its effect was drastically reduced about that. They proposed an empirical correlation for HTC as a less-than-linear function of \( R_a \), i.e., \( h \propto R_a^m \), where \( m \) is also a function of the contact angle.
Kim et al. [5] also reported enhanced HTC on roughened surface due to increased active nucleation site density. However, they observed that CHF is strongly enhanced by surface roughness, which seems to be in contrast with the conclusions drawn in Refs. [1-3].

Fan et al. [6] reported surface roughness effects on HTC and CHF similar to previous studies. Rougher surfaces exhibit a larger number of nucleation sites, smaller bubbles, higher departure frequency, and higher HTC. Instead, CHF does not depend on roughness as much as on wettability. Recently, Paz et al. [7] have investigated the effect of surface features on subcooled flow boiling and concluded that an averaged 1-D parameter is not representative of the 3-D surface morphology. Yet, they proposed a heat flux enhancement factor incorporating the surface average roughness ($S_a$) parameter. Paz et al. [8, 9] examined the effects of surface roughness on bubble size distribution and nucleation site density by post-processing HSV images. Rougher surfaces have smaller bubbles and a narrow range of diameters, because rough structures impede the development of bubble growth. Rougher surfaces have higher nucleation site density in the fully developed boiling regime, because of the cavity activation that occurs when the bubble grow on the surface (and the bubble microlayer dries out).

O’Hanley et al. [10] examined the separate effects of surface wettability, roughness, and porosity on pool boiling CHF using non-metallic engineered surfaces. They explore a wide range of roughness values, and demonstrated that roughness per se does not affect pool boiling CHF. Their results corroborate the arguments made in several pool boiling studies on metallic heaters [1-3, 6] that roughness is not a clear indicator of CHF.

In summary, even though there is a general consensus that surface parameters such as wettability and roughness are not unique indicators of the boiling performance, still, very often these are the sole parameters use to correlate CHF or HTC with the surface properties.
1.2.2 Effects of Substrate Thickness on Critical Heat Flux

Substrate thickness is known to affect the CHF limit in pool boiling conditions [12-21]. When the heating wall is thick enough, CHF is not affected by the wall thickness and has constant value, the so-called “asymptotic” CHF. On the other hand, when wall thickness is thin enough to affect the energy transport within the substrate, the CHF decreases as the wall thickness decreases. The asymptotic thickness is defined as the thickness at which the CHF limit is 90% of its asymptotic value. The asymptotic thickness was observed to depend on the effusivity of the substrate material (Figure 1.3). Materials with higher effusivity transfer energy better than materials with low effusivity, and have a smaller asymptotic thickness, i.e., one can reach the asymptotic CHF value with thinner samples compared to materials with low effusivity.

There are plenty of empirical correlations (e.g., Eqs. (1.1) throughout (1.5)) to predict the asymptotic thickness, \( \delta_{asy} \), or how the CHF depend on the sample thickness based on the material effusivity in saturated pool boiling conditions:

- Guglielmini and Nannei (1976) [15]
  \[
  \delta_{asy} = 2.35 \cdot 10^{-4} \left( \frac{\varepsilon}{10^4} \right)^{-3.26}
  \tag{1.1}
  \]

- Bar-Cohen and McNeil (1992) [16]
  \[
  \frac{q''_{CHF}}{q''_{CHF,asy}} = \frac{\delta \varepsilon}{\delta \varepsilon + C} \text{ C<1}
  \tag{1.2}
  \]

- Golobic and Bergles (1997) [17]
  \[
  \delta_{asy} = 8 \cdot 10^{-4} - 3.5 \cdot 10^{-8} \varepsilon \text{ (for } \varepsilon < 22) \tag{1.3}
  \\
  \delta_{asy} = 4.5 \cdot 10^{-4} - 8 \cdot 10^{-10} \varepsilon \text{ (for } \varepsilon > 22) \tag{1.4}
  \]
  \[
  \frac{q''_{CHF}}{q''_{CHF,asy}} = 1 - e^{-\left(\frac{\delta \varepsilon}{2.44}\right)^{0.8498} - \left(\frac{\delta \varepsilon}{2.44}\right)^{0.0581}}
  \tag{1.5}
  \]

where \( \delta \) is the substrate thickness in mm, and \( \varepsilon \) is material effusivity in kW√s/(m²K). Table 1.1 summarizes the experimental conditions (e.g., working fluid, heater geometry, and heater material) on which these correlations were developed. Note that not all the studies in Table 1.1 produced a correlation.
Table 1.1 Literature review of investigations studying pool boiling asymptotic CHF and heater thickness

<table>
<thead>
<tr>
<th>Author</th>
<th>Year</th>
<th>Fluid</th>
<th>Geometry</th>
<th>Material (# data)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Carne and Charlesworth</td>
<td>1966</td>
<td>n-propanol</td>
<td>Horizontal plate (7×76 mm)</td>
<td>Copper (5)</td>
</tr>
<tr>
<td>[12]</td>
<td></td>
<td></td>
<td></td>
<td>Steel (6)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Monel (4)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Inconel (4)</td>
</tr>
<tr>
<td>Tachibana and Akiyama</td>
<td>1967</td>
<td>Water</td>
<td>Horizontal plate (8×55 mm)</td>
<td>Copper (1)</td>
</tr>
<tr>
<td>[13]</td>
<td></td>
<td></td>
<td></td>
<td>Aluminum (3)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Nickel (2)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>SS (9)</td>
</tr>
<tr>
<td>Guglielmini and Nannei</td>
<td>1976</td>
<td>Water</td>
<td>Horizontal cylinder (OD 10 mm), Electro deposition of thin (0.4~0.7 μm) metal layers</td>
<td>Copper (30)</td>
</tr>
<tr>
<td>[15]</td>
<td></td>
<td></td>
<td></td>
<td>Zinc (33)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Nickel (41)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Tin (30)</td>
</tr>
<tr>
<td>Bar-Cohen and McNeil</td>
<td>1992</td>
<td>Dielectric liquid</td>
<td>Horizontal plate</td>
<td>Pt</td>
</tr>
<tr>
<td>[16]</td>
<td></td>
<td></td>
<td></td>
<td>Silicon</td>
</tr>
<tr>
<td>Golobic and Bargles</td>
<td>1997</td>
<td>FC-72</td>
<td>Horizontal plate (5×50 mm)</td>
<td>Copper (4)</td>
</tr>
<tr>
<td>[17]</td>
<td></td>
<td></td>
<td></td>
<td>Aluminum (4)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Brass (11)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Steel (9)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>SS (6)</td>
</tr>
</tbody>
</table>

Figure 1.3 shows how the effusivity of several materials change with temperature. Copper has the highest effusivity, whereas Stainless Steel (SS) and Inconel have the lowest effusivity among materials used in the previous pool boiling studies. Figure 1.4 shows CHF data and correlations for Copper, SS, and Inconel. Asymptotic thickness values for Copper are scattered, range between 0.015 mm and 0.0035 mm. Whereas, asymptotic thickness values for SS show good agreement among different authors, i.e., 0.55 mm. This observation suggests that the existing empirical correlations, Eqs. (1.1) throughout (1.5), which have been extensively used in recent years, predict
the behavior of materials with low effusivity reasonably well.

In subcooled flow boiling conditions, Del Valle M. [21] developed an empirical correlation, Eq (1.6), to predict the normalized CHF for SS as a function of the sample thickness only. The recommended asymptotic thickness value recommended by these authors was 0.5 mm.

- Del Valle M. (1983) [21]

\[
\frac{q^{''}_{\text{CHF}}}{q^{''}_{\text{CHF,asy}}} = 1 - e^{-12\delta}
\]

(1.6)

Although Zircaloy-4 and SS have similar effusivity (see Figure 1.3), there are no experimental data for Zircaloy-4 in flow boiling conditions. Note that, to fill this gap, we conducted subcooled flow boiling CHF experiments for fresh Zircaloy-4 heaters with different thickness (see Section 5.2).

Figure 1.3 Effusivity of several materials of interest as function of temperature
Figure 1.4 Pool boiling CHF data and empirical correlations for Copper (up) and SS and Inconel (bottom)
1.2.3 Effects of Zircaloy-4 Oxidation on Boiling Heat Transfer

1.2.3.1 Oxidation kinetics

Zirconium alloys are used as nuclear fuel cladding material in light water reactors because of their good mechanical properties (e.g., high hardness and ductility) and low thermal neutron absorption cross-section. Traditionally, Zircaloy-4 claddings are used in PWRs. Zircaloy-4 claddings undergo significant oxidation during in-pile operation due to the high operating temperature (~350 °C for PWR) and the chemicals dissolved in the coolant. This oxidation causes changes in the surface morphology and wettability.

When the cladding surface oxidizes, zirconium oxide, i.e., zirconia (ZrO$_2$), forms on the top of the surface, according to the reaction:

$$Zr + 2O^{2-} = ZrO_2 + 2Vo + 4e^-$$

where Vo indicates a lattice vacancy in the ZrO$_2$ layer [22]. Zirconium oxide has distinct characteristic features depending on the oxidation stage. Oxide weight gain in zirconium alloys reveals pre-transition, transition, and post-transition stages, as shown in Figure 1.5. The weight gain can be correlated with the oxide layer thickness: a 14.9 mg/dm$^2$ of weight gain corresponds to ~1 μm of oxide thickness [23].

![Diagram of Zirconium Oxide Growth Kinetics](image)

**Figure 1.5** Schematic drawing for zirconium oxide growth kinetics [23].
The pre-transition stage is characterized by a sub-linear growth rate. The oxidized layer is thin, black, protective, and tightly adherent to the Zircaloy substrate [22]. The oxide weight gain, \( \Delta W \), follows the power law:

\[
(\Delta W)^n = kt
\]

where \( k \) is the pre-exponential factor and \( t \) is time. Two commonly-used correlations for Zircaloy-4 weight gain during steam oxidation were developed by Baker and Just (B-J) [24], and Cathcart and Pawel (C-P) [25], respectively:

\[
\text{B-J: } (\Delta W)^2 = 33.3 \times 10^6 \ t \exp \left( -\frac{45500}{RT} \right) \text{ (mg/cm}^2\text{)}^2
\]

\[
\text{C-P: } (\Delta W)^2 = 1.471 \times 10^6 \ t \exp \left( -\frac{39940}{RT} \right) \text{ (mg/cm}^2\text{)}^2
\]

where \( t \) is time in seconds, \( T \) is temperature in K, and \( R \) is the universal gas constant, 1.987 cal/K/mol (or 8.314 J/K/mol). Increasing the temperature causes higher weight gain rates (i.e., faster oxidation). This is because temperature increases the chemical reaction kinetics for both the oxide layer formation and volatilization, and it also affects the fluid properties, e.g., the diffusion coefficients [26].

The transitory stage lies between the pre- and post-transition stages and is characterized by a cyclic sublinear growth rate [27]. Bryner [28] showed that this transitory regime consists of a series of successive stages. This regime results from the superposition of multiple pre-transition growths that are slightly out of phase with each other. The transition to a new oxidation cycle is triggered by the destabilization of the oxide layer. The oxide layer cracks as a result of the accumulation of high compressive stresses due to the zirconia volume expansion. The transformation of initially tetragonal ZrO\(_2\) to the monoclinic system results in a volume increase. The porosity formed in the oxide facilitates the percolation of the coolant into the underlying metal. Consequently, the passivating oxide layer flakes off, exposing new, unoxidized surfaces to the coolant. Then, a new oxidation cycle starts [22, 27, 28].

Finally, the post-transition stage is characterized by a linear growth rate, and the oxide is white and non-protective [29].

During the in-core operation, the cladding of the fuel rod undergoes these three phases of oxidation.
The thickness of the oxide layer is expected to have grown to up to several tens of microns at the end of the cladding life, as shown Figure 1.6. In order to duplicate oxide layers formed during in-pile condition, in this study, we have grown oxide layers up to ~20 μm thick. Details are discussed in Section 5.3.

![Figure 1.6](image)

**Figure 1.6** Oxide thicknesses as a function of burnup in 900 MW PWRs (Figure adapted from Ref. [30]).

### 1.2.3.2 Boiling experiments on the effect of Zircaloy oxidation

The typical test protocol of previous pool boiling studies on oxidized Zircaloy-4 is shown in Figure 1.7 (a). One sample was set to be the unoxidized reference sample, and other multiple samples were oxidized for short-term to examine the effects of pre-transition oxide on pool boiling CHF. Previous saturated pool boiling studies concluded that CHF is enhanced by the pre-transition oxide due to increased wettability [31] or increased roughness [32]. However, this approach seems questionable considering the CHF and HTC variability that we observed on multiple, unoxidized and nominally identical samples, discussed in Section 1.1. Different fresh (i.e., unoxidized) surfaces may have different bubble dynamics, HTC, or CHF in subcooled flow boiling.

In order to eliminate uncertainties arising from the randomness of the fresh surface finish, a new testing protocol (Figure 1.7 (b)) is proposed in this study, using the same exact sample throughout the entire test campaign. Note that boiling (including CHF) does not seem to change the surface microscopic characteristics (see details in Appendix A). Also, as explained in Section 1.2.3.1, the scope of the current study includes both short-term and long-term oxidation stages to mimic the
in-core Zircaloy-4 aging process. In order to systematically analyze the evolution of the surface properties (up to the long-term, post-transition oxide), we characterized not only how the surface wettability changes, but also the micro- and nano-scale topography, statistical surface parameters (e.g., $S_a, r$), and porosity (see Section 5.3.2). Finally, previous studies failed to obtain conclusive results to explain the mechanisms enhancing CHF in pool boiling. This is possibly due to lack of high-resolution diagnostic tools to examine the bubble dynamics, i.e., to observe what exactly happens on the boiling surface. In this work, ad hoc HSV imaging and quantitative analysis of bubble dynamics were used (see Section 5.3.3) to investigate the separate effect of Zircaloy-4 oxidation on subcooled flow boiling heat transfer. In fact, fundamental studies on the effects of surface oxidation on CHF leading mechanisms in subcooled flow boiling are important to filling the missing knowledge gap for further modeling of safety margins in nuclear power plants [33].

Figure 1.7 Test protocol of (a) previous studies and (b) the current study.
1.2.4 Effects of Accident Tolerant Fuel Cladding Material on Boiling Heat Transfer

After the Fukushima Daiichi nuclear accident in 2011, accident tolerant fuel (ATF) cladding materials have received great attention due to their resistance to oxidation and generation of hydrogen under the high-temperature steam environment expected in certain accident conditions [34-52]. The cladding material is one of the important factors that affect boiling heat transfer and CHF in LWRs. Different materials may have different thermal hydraulic properties such as wettability and effusivity. In this study, the performance of Cr-coated Zircaloy-4 and Fe-21Cr-5Al Alloy (Kanthal APMT) was tested in subcooled flow boiling.

Although many pool boiling CHF experiments have been conducted on Cr-coated surfaces, it is difficult to compare how coatings affect the surface properties (e.g., wettability, roughness) and the boiling behavior (CHF, HTC). The data of previous studies are scattered and not in good agreement with each other [36-37]. Crucially, different coating techniques (e.g., sputtering, cold spray, electroplating) may generate different types of Cr-coated surface [36, 38]. Table 1.2 summarized conditions and results of bare and Cr-coated samples testing in saturated pool boiling. According to these studies, Cr-coating by electroplating and cold spray makes the surface more hydrophobic and reduces CHF. On the contrary, Cr-coating by DC sputtering makes the surface super-hydrophilic and enhances CHF. In general, the average roughness becomes slightly higher, but very similar to the bare surface.

Umretiya et al. [36] reported slightly increased average surface roughness and decreased contact angle on Cr-sputtered surface. Nano-scale topography of Cr-sputtered surface showed that the asperities have rounded shapes. The sub-micron features were presumed to promote wicking and cause additional wetting of a water droplet, which resulted in higher wettability on Cr-coated surface.

Lee et al. [38] conducted subcooled flow boiling experiments on a Zircaloy-4 rod and two Cr-coated Zircaloy-4 rods, one coated by PVD sputtering and the other by cold spray. The cold spray Cr-coated surface has higher roughness compared to both fresh Zircaloy-4 and PVD sputtered Cr-coated surfaces, which induced smaller and faster bubble nucleation. Accordingly, the cold spray Cr-coated Zircaloy-4 surface leads to a higher void fraction, higher HTC, and lower CHF than the
other surfaces. Instead, the PVD sputtered Cr-coated Zircaloy-4 showed a boiling behavior (HTC and CHF) similar to the fresh Zircaloy-4 cladding.

**Table 1.2** Literature review for saturated pool boiling CHF on Cr-coating

<table>
<thead>
<tr>
<th>Author</th>
<th>Kam et al. [39]</th>
<th>Jo et al. [37]</th>
<th>Son et al. [40]</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Year</strong></td>
<td>2015</td>
<td>2019</td>
<td>2020</td>
</tr>
<tr>
<td><strong>Bare surface</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Material</td>
<td>SS</td>
<td>ZIRLO</td>
<td>SS304</td>
</tr>
<tr>
<td>Thickness (mm)</td>
<td>1.2</td>
<td>0.8</td>
<td>2</td>
</tr>
<tr>
<td>Contact angle (°)</td>
<td>61±5</td>
<td>64±3</td>
<td>63±5</td>
</tr>
<tr>
<td>Average roughness, Rₐ (nm)</td>
<td>25.44±2.32</td>
<td>120</td>
<td>111±22**</td>
</tr>
<tr>
<td>CHF (kW/m²)</td>
<td>1020</td>
<td>645</td>
<td>700</td>
</tr>
<tr>
<td><strong>Cr-coating</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Coating method</td>
<td>Electroplating</td>
<td>Cold spray</td>
<td>DC sputtering***</td>
</tr>
<tr>
<td>Coating thickness (μm)</td>
<td>1</td>
<td>64~79</td>
<td>1.5</td>
</tr>
<tr>
<td>Contact angle (°)</td>
<td>81±4</td>
<td>77±6</td>
<td>0</td>
</tr>
<tr>
<td>Average roughness, Rₐ (nm)</td>
<td>27.61±0.76</td>
<td>74*</td>
<td>123±10 Similiar</td>
</tr>
<tr>
<td>CHF (kW/m²)</td>
<td>660</td>
<td>571</td>
<td>1015</td>
</tr>
<tr>
<td>Boiling HTC</td>
<td>Reduced</td>
<td>Reduced</td>
<td>Enhanced</td>
</tr>
<tr>
<td>Bubble size</td>
<td>Larger</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*Polished with 600-grit sandpaper after deposition.

**Selected case polished with 800-grit sandpaper.

**Selected case sputtered at room temperature.

Testing conditions and results for reference samples and FeCrAl or FeCrAl-coated samples tested in saturated pool boiling are summarized in Table 1.3
<table>
<thead>
<tr>
<th>Author</th>
<th>Ali et al. [41]</th>
<th>Jo et al. [37]</th>
<th>Seo et al. [42]</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Year</strong></td>
<td>2018</td>
<td>2019</td>
<td>2016</td>
</tr>
</tbody>
</table>

| **Material**    | Zircaloy-4      | ZIRLO          | SS316           |
| **Thickness (mm)** | -               | 0.8            | 2               |
| **Contact angle (°)**  | 74.56±3         | 64±3           | 84.8            |
| **Average roughness, R<sub>a</sub> (nm)**  | 358±29          | 120            | 103             |
| **CHF (kW/m<sup>2</sup>)**  | 784             | 645            | 1047±119        |

| **Coating method**  | Fe-13Cr-5Al Alloy | Cold spray | DC sputtering** |
| **Coating thickness (µm)**  | -               | 40          | 1               |
| **Contact angle (°)**  | 75.48±3          | 89±9        | 79.9            |
| **Similar** | **Less wetting** | **More wetting** |
| **Average roughness, R<sub>a</sub> (nm)**  | 590±47          | 67*         | 133             |
| **Rougher** | **Similar** | **Reduced** | **Enhanced** |
| **CHF (kW/m<sup>2</sup>)**  | 807             | 630          | 1258±80         |
| **Boiling HTC**  | **Enhanced**    | -            | **Enhanced**    |
| **Bubble size**  | -               | -            | -               |

*Polished with 600-grits sandpaper after deposition.
** Selected case sputtered at room temperature.

Although wettability and CHF do not have consistent trends, the HTC seems to be enhanced on FeCrAl. The similar behavior was observed in subcooled flow boiling experiments. Lee et al. [43] conducted steady and transient internal-flow subcooled flow boiling CHF tests on Zircaloy-4, Inconel 600, and Fe-13Cr-6Al Alloy tubes. They reported higher average roughness (R<sub>a</sub>:0.43±0.01 µm) and higher wettability (contact angle: 69.43±1.26 °) on FeCrAl compared to Zircaloy-4 (R<sub>a</sub>:0.24±0.02 µm, contact angle: 76.48±1.19 °). The surface morphology and heating wall thickness of FeCrAl were different from Zircaloy-4. Enhanced HTC and CHF on FeCrAl were observed. However, the single-phase regime of the boiling curves shows big deviations.
among the different materials (although the authors claimed to run the experiments in the same flow conditions), which makes the comparison of CHF results less convincing.

In this work, we developed a testing protocol to separate surface effects due to ATF coatings or materials, as much as possible (see Figure 1.8).

First, as-received Zircaloy-4 and FeCrAl samples were simultaneously and uni-directionally polished to minimize uncertainties arising from random scratches. Then, in order to separate the effect of Cr-coating from intrinsic characteristics of different samples, Cr was sputtered on the fresh Zircaloy-4 and FeCrAl samples. Boiling tests were conducted with both fresh and coated samples, as indicated in Figure 1.8. Since boiling is affected by both surface conditions and thermal-properties of the substrate material, Cr-coating on different substrate materials would allow separating surface effects from material properties. Last but not least, previous ATF studies did not enable high-resolution visualizations of the boiling process, which is useful to understand how the bubble dynamics and corresponding heat transfer mechanisms change due to ATF coatings. In this work, we imaged the boiling process and extracted fundamental boiling parameters from high-resolution HSV images (see Section 5.4.2).

Figure 1.8 ATF test protocol of the current study.
1.2.5 High-Speed Video (HSV) Analysis of the Boiling Process

In recent years, the boiling community has put in a great deal of efforts to closely examine the near-wall boiling dynamics. Infrared (IR) thermometry [53-54] or phase detection [55] techniques, which enable visualizations of bubble interactions on the boiling surface, can be used to this end. However, to work, these techniques require a transparent substrate and a thin visible-light transparent and IR-opaque layer on the boiling surface. Thus, they cannot be used to study the boiling process on thick metallic samples, e.g., nuclear reactor fuel claddings. With thick metallic substrates, temperature fields can be only measured at the backside of the heater, and, consequently, losing information of bubble interaction process on the boiling surface.

Due to these limitations, HSV shadowgraphy is still the best way to image the bubble dynamics on these thick metallic heaters.

The majority of previous studies using HSV in subcooled flow boiling has focused on the behavior of single bubbles at low heat flux conditions, using both front and lateral views (e.g., see Refs. [56-69]), as shown in Figure 1.9. By frame-by-frame observation, scientists could obtain bubble grow parameters (e.g., departure frequency, diameter, sliding length).

![Figure 1.9](image)

**Figure 1.9** Typical HSV imaging of single bubble dynamics in the literature: (a) examples of back-lit lateral view (adapted from Ref. [60]) and (b) front view (adapted from Ref. [65]).
However, when the heat flux increases, this approach is unpractical, because of the complicated boiling dynamics, including bubble interactions, and the overwhelming number of bubbles, as shown in Figure 1.10.

![Figure 1.10](image)

**Figure 1.10** Front-lit, front-view HSV image of the boiling process from the current study.

Note that, for a non-smooth metallic surface, the nucleation site density can reach values as high as 100 to 500 sites/cm² even at relatively low heat fluxes (i.e., far from the CHF). Therefore, there is a need for automated bubble motion analysis tools to quantify the desired boiling parameters from high-speed video images. Moreover, crucial boiling parameters such as the nucleation site density cannot be measured from lateral views, except at very low heat flux where only a few bubbles nucleate on the heated surface.

In several studies, the total number of active nucleation sites or the number of bubbles over a heated surface was obtained using front view HSV imaging, but only up to relatively low heat fluxes and by manual detection [70-71]. Meanwhile, efforts to automatically recognize multiple bubbles from HSV imaging in horizontal subcooled flow boiling using image-processing or machine vision techniques have been made [8, 72-75].

Ideally, front view images can be used to measure nucleation site density, bubble departure diameter and departure frequency, and get estimates of the void fraction in the flow. In subcooled flow boiling, when a bubble leaves the surface, it starts to condense as soon as it escapes from the near-wall superheated layer. There are plenty of bubbles like this, which are captured in the videos.
However, detached bubbles should not be counted in the wall heat transfer analysis. Still, it is hard to determine whether a bubble is attached or detached from the heating surface based on front-view images (see Figure 1.10), because we cannot directly observe the bubble triple contact line. Being able to determine the exact moment of detachment is critical to produce accurate bubble departure diameter and growth time measurements, and also to distinguish the moment of departure from the nucleation site from the moment where the bubble ultimately departs from the surface (after sliding on it).

Standardized bubble segmentation techniques using image-processing or machine vision work relatively well only under certain circumstances, e.g., large bubble size, spherical or elliptical bubble shape, good contrast between the bubble and the background (i.e., the heating wall), or a bubble having the brightest illumination spot. These conditions are not met in many cases, e.g., when flow boiling experiments are conducted on surfaces with different finish or brightness, with a non-homogeneous lighting environment, and in operating conditions (e.g., high heat flux, high mass flux, high pressure) where bubbles can be very small or have irregular shape. For example, in Figure 1.10, bubbles have a similar contrast and texture as the background and the shape of a single bubble is disturbed by the turbulent wakes created by the surrounding bubble motion (e.g., sliding or coalescence). The methodologies introduced in the literature do not allow to segment bubbles from this kind of HSV images.

To overcome this limitation, in this work, we developed a methodology that include machine learning and optical flow techniques to post-process HSV images and extract the complete set of fundamental boiling parameters (e.g., location and density of nucleation site, as well as departure diameter, growth time, departure frequency, lift-off diameter, sliding time, and sliding length per each nucleation site) necessary to model the partitioning of the heat fluxes at the boiling surface. The proposed HSV post-processing system consists of bubble segmentation, bubble tracking, and bubble life-cycle identification, and are validated by comparing the quantified parameters to the measurements performed by IR thermometry using a special heater, which allows simultaneous access to both near-wall boiling (i.e., temperature and heat flux fields) and vapor bubble parameters (see Section 3.2).
1.3 Scope of Work

The tasks completed to achieve the goals of the present study are summarized hereafter:

- **TASK #1.** The separate effect of surface morphology on subcooled flow boiling was investigated. In most of previous boiling studies using nuclear fuel cladding material, researchers had prepared metallic samples by polishing the boiling surface either randomly or uni-directionally. In this study, we examined how groove direction affects subcooled flow boiling.

- **TASK #2.** The effect of Zircaloy-4 substrate thickness on subcooled flow boiling CHF was investigated to prepare a sample within the “safe” thickness range, i.e., within a range of thickness that does not affect the CHF value.

- **TASK #3.** The separate effect of Zircaloy-4 oxidation on subcooled flow boiling was investigated. To that end, the exact same sample in fresh, pre-transition (short-term) oxidation, transition oxidation, and post-transition (long-term) oxidation conditions was tested.

- **TASK #4.** The separate effect of ATF cladding material on subcooled flow boiling was investigated. Fresh Zircaloy-4 and fresh FeCrAl samples were prepared by carefully controlling the surface finish conditions. After conducting boiling tests on fresh samples, Cr was coated on each surface to examine the separate effect of Cr-coating on subcooled flow boiling.

- **TASK #5.** In order to accomplish TASKs #1 throughout #4, a systematic heater fabrication procedure was developed to control surface finish conditions throughout the whole process, e.g., polishing, oxidation, sputtering, and boiling.

- **TASK #6.** A surface characterization technique was established. Micro- and nano- scale topography and morphology were measured on the exact same region of the exact same sample before (i.e., fresh) and after any surface modification. This technique enables to complete separate effect studies by characterizing how surface features affect the boiling behavior.

- **TASK #7.** Subcooled flow boiling experiments were carried out using high-resolution high-speed video imaging, by following a well-established protocol to control the operating conditions.
• TASK #8. Post-processing methodologies for IR and HSV images were developed. A
STAR-CCM+ 3-D inverse conduction model as well as a MATLAB 1-D inverse
conduction algorithm were developed to obtain wall superheat and heat flux (i.e., boiling
curve) from IR data. A machine learning tool and an optical flow algorithm were
implemented to post-process the HSV images and extract quantitative fundamental boiling
parameters.
• TASK #9. Bubble dynamics were quantitatively analyzed and incorporated into a heat flux
partitioning model to understand how surface conditions affect the boiling heat transfer
mechanisms.
Chapter 2

Experimental Set-up and Methodology

2.1 Metallic Heater Design

A Zircaloy-4 heater is designed to mimic the nuclear fuel cladding, as illustrated in Figure 2.1. The heat released by Joule effect on one side of the heater is delivered to the other side (in contact with water) by conductive heat transfer.

![Figure 2.1 Conceptual diagram of a metallic heater (side view).](image)

The typical heater size is $20 \times 20 \times 1 \text{ mm}^3$ and the active heating area is $10 \times 10 \text{ mm}^2$ (see Figure 2.2). The boiling surface is polished to have the similar average roughness as a commercial fuel cladding, i.e., $R_a: 0.2 \sim 0.3 \mu m$. The airside surface is mirror-polished and coated with multiple thin layers, as shown in Figure 2.2. The first layer is a $4 \mu m$ thick $\text{Al}_2\text{O}_3$ insulation layer. The second layer is a $1 \mu m$ thick Joule-heating $	ext{Cr}$ layer. Finally, $0.1 \mu m$ thick, Ag electro-pads are coated on the side of the chromium layer and serve as electrical connections. They limit the active heating area to a $10 \times 10 \text{ mm}^2$ square. Thin Cr film ($\sim 20 \text{ nm}$) is deposited between the Cr heating
layer and the Ag pads to strengthen the mechanical bonding. The infrared radiation emitted by the chromium layer is used to measure the temperature at the airside of the metallic substrate and, after the appropriate post-processing steps, the temperature and heat flux on the boiling surface (see details in Section 2.3.1 and Section 3.1).

This heater design is applicable to any type of material, as depicted in Figure 2.1 (right). For the FeCrAl samples, due to the poor bonding between FeCrAl and Al₂O₃, a ~20 nm Cr bonding layer was deposited between the metallic substrate and the Al₂O₃ coating.

**Figure 2.2** Multilayer heater design (not to scale).
2.2 Heater Fabrication

A systematic heater fabrication procedure was developed to carefully control surface finish conditions during polishing, oxidation, and boiling (see Figure 2.3). Each step is necessary to preserve the heater quality throughout the oxidation and testing process.

![Figure 2.3 Heater fabrication procedure.](image-url)
The boiling side of a 20 × 20 × 1 mm³ as-received Zircaloy-4 sample is randomly polished with a Buehler CarbiMet™ 240-grit sandpaper at 20 N, 200 rpm for 2~3 minutes with a Buehler AutoMet™ 250 polisher to create average roughness of 0.2~0.3 μm. The airside of the same sample is mirror-polished by gradually increasing the grit number (e.g., 240, 400, Microcut® SiC 800, Microcut® SiC 2000) and using MetaDi™ Polycrystalline Diamond Suspensions (with a diamond size of 3 μm, 1 μm and 0.05 μm). This is an important step because imperfections on the airside surface may result in failure of Al₂O₃ insulation layer. After polishing, the sample is sonicated in Acetone and then rinsed with Ethanol, Isopropanol, and DI water to remove residual Crystalbond and debris.

The layers described in Section 2.1 are coated on the mirror-polished surface by physical vapor deposition (PVD) using a Kurt J. Lesker PRO Line PVD75 at 3 mT with Argon flow (base pressure: 2E-6 Torr). During the deposition, the substrate rotates at 8 rpm at room temperature. In order to enhance the bonding between Al₂O₃ and the Zircaloy-4 substrate, a non-reactive 0.1 μm thick Al₂O₃ is first deposited at a rate of ~0.023 Å/s. Then, a 4 μm thick Al₂O₃ layer is coated by reactive sputtering at a rate of ~0.8 Å/s at 3 mT with a 11% Oxygen, 89% Argon flow. Slower deposition rate would enhance the quality of Al₂O₃ insulation layer.

The subcooled flow boiling experiment is conducted on the fresh sample (Section 2.3).

Boiling surface of the fresh sample is fully characterized (see Section 2.4). Note that, boiling (including CHF) does not change the surface structures (details of this investigation can be found in Appendix A).

The heater layers (i.e., on the airside) are polished out before oxidation because the presence of the coatings, especially the Al₂O₃ layer, prevent the airside surface from oxidizing, and this can cause a deformation of the sample due to non-symmetric volume expansion. When polishing the airside, a prop is positioned underneath the sample to prevent the contact between the boiling surface and the sample holder plate. A prop has a square hole larger than active boiling area, 10 × 10 mm². It is crucial to avoid any damage to the boiling surface during the whole procedure to preserve the separate effect investigation protocol.

The sample is oxidized in 500 °C humid air flow in a tube furnace. The tube furnace is heated up to the prescribed temperature set point (500 °C). The rate of temperature increase (or decrease) is limited to 5 °C/min to minimize thermal stresses. When the temperature of the furnace reaches the prescribed value, synthetic air (AI B300) is sparged through a water flask
at room temperature. Then, the humid air is delivered to the tube furnace with the controlled volumetric flow rate.

7 Oxidation may change the surface morphology. Therefore, the airside of the sample is mirror-polished again after the oxidation process to successfully re-deposit the heater layers.

8 The multilayer heating elements are coated on the mirror-polished surface for the oxidized sample.

9 Subcooled flow boiling experiment is conducted on the oxidized sample.

10 Boiling surface of the oxidized sample is fully characterized.

Step 5~10 are repeated several times until the sample oxide reaches the post-transition stage.

Note that the sample thickness is carefully controlled at every polishing step to verify that the sample thickness is larger than the asymptotic thickness limit (see Section 5.2).

For morphology effect investigations (see Section 5.1), step 6 and 7 were skipped. For ATF cladding material effect investigation (see Section 5.4), Cr was coated on the boiling surface at step 6 instead of oxidizing the surface.
2.3 Flow Boiling Facilities and Diagnostics

A picture of the flow loop is shown in Figure 2.4. The flow loop is built using 316 stainless steel fittings and tubing, and includes a variable speed pump, a pressurizer, a heat exchanger, a membrane filter, a degassing filter, and heat tape pre-heaters.

![Figure 2.4 Picture of the flow loop.](image)

The loop is controlled through a LabVIEW interface, through which we can regulate the loop pressure, the flow rate (and, accordingly, the Reynolds number using temperature-dependent properties), the temperature at the inlet of the test section, and the heat released at the boiling surface.

Before running boiling experiments, non-condensable gases are removed circulating the fluid through a secondary loop. This secondary loop has a degassing filter with a membrane which is permeable to non-condensable gases, but impermeable to liquids. The outer side of the membrane, i.e., the side not in contact with the fluid, is connected to a vacuum pump that is used to pull the vacuum. When the vacuum is pulled, the non-condensable gases dissolved in the liquid cross the
membrane. This secondary loop has also another type of membrane filter, which is used to trap and remove dust particles and other kind of debris from the working fluid. The shell-and-tube type heat exchanger (HX) is connected to the building processing water supply. Two preheaters are designed to compensate the amount of heat removal from the HX and control the energy balance of the loop. This energy balance also includes the power applied to the Zircaloy-4 heater. The LabVIEW interface is used to control the operation, but also to record all the data from the pressure and temperature sensors available in the loop, as well as the current and voltage circulating through the heater. In order to capture departure from nucleate boiling (DNB) without damaging boiling surface, we cut the power immediately when an irreversible dry spot starts to grow (i.e., we minimize the time spent in post-CHF conditions). The growth of an irreversible dry spot can be observed by drastic increase in local IR counts using a IR camera (see Figure 2.6). In this way, heater burnout or any alteration can be prevented when CHF occurs, so that a given sample can be re-used for the next test. A heater is installed in a Shapal™ ceramic cartridge, as shown in Figure 2.5. The heater cartridge provides mechanical support and electrical insulation for a heater. The flow channel cross section area is $1 \times 3 \text{ cm}^2$.

![Figure 2.5 Heater cartridge.](image)

The heater cartridge is part of the test section, which is placed at the upper part of the flow channel (where the fluid flows upward). The position of the test section is such that the flow is fully
developed when it flows on top of the heater (see Figure 2.4).

The configuration of optical diagnostics is shown in Figure 2.6. The IR camera (IRC806HS) measures the IR radiation from the active heating area on the airside of the heater with a resolution of 115 $\mu m$/pixel at 2500 frames per second (fps). The 3-D IR counts distributions are converted into 3-D temperature distributions using an ad-hoc calibration curve that correlates counts to temperature. As shown in Figure 2.6 (a), the temperature distribution is rather uniform in the center region of the active heating area. This 3-D temperature field is used as input to an inverse conduction model (described in Section 3.1) which we implemented in STAR-CCM+ to compute the average temperature and heat flux on the boiling surface. The HSV camera (Phantom® v2512) captures the bubble dynamics on the waterside of the heater with a resolution of 30 $\mu m$/pixel at 10000 fps. A white LED light is used to lit the bubbles from the front.

![Figure 2.6](image)

**Figure 2.6** Configuration of optical diagnostics. (a) Schematic diagram (b) Real picture.

In subcooled flow boiling, the wall superheat highly depends on the liquid subcooling. Figure 2.7 shows measurements of the test section inlet temperature as a function of time after the inlet temperature has reached a setpoint temperature of 90 °C (or 363.13 K, i.e., 10 K subcooling). Note that there is initial overshoot of ~2 °C. After that, the inlet temperature fluctuates within a ±2 °C
range and becomes stable only after a few hours due to an inherent stabilizing function in the temperature controller system. The length of this stabilizing time is critical to get accurate and reliable results. For example, Figure 2.8 shows two boiling curves obtained at different stabilizing time, together with measurements of inlet temperature, pressure and mass flux. One test (red circles) is conducted immediately when the inlet temperature reaches 90 °C for the first time (i.e., after a stabilizing time of 0 h). The other test (teal circles) is conducted after a stabilizing time of 2 h. Note that, since the two tests were conducted on different fresh Zircaloy-4 samples, the HTC and CHF cannot be directly compared. However, it can clearly be seen that in the 0 h case, the non-regular trend of the boiling curve (e.g., the spike marked with a red circle) is due to an oscillation of the subcooling temperature at the inlet of the test section. Therefore, it is critical to keep the operating conditions (especially inlet subcooling) very stable throughout the duration of an experiment. In the current study, the fluctuations of inlet subcooling is kept within ±1 K, i.e., we stabilized the inlet temperature for 30 to 60 minutes before running the experiment.

![Figure 2.7 Inlet temperature profile as a function of time.](image)

![Figure 2.8 Operating conditions. From left to right: boiling curve, inlet subcooling, pressure, and mass flux.](image)
2.4 Surface Characterization

In the current study, the micro- and nano-scale topography and morphology of a surface were characterized before and after surface modifications on the same exact surface region. This expedient is important to eliminate characterization uncertainties coming from randomly selected surface regions. Even on one given sample, different locations may have different micro- or nano-scale features. This uncertainty becomes critical for a randomly polished sample. In order to elucidate the separate effect of surface oxidation or Cr-coating, each surface was characterized in three steps at increasing resolution.

The first step consists in measuring the micro-scale surface topography, as shown in Figure 2.9 (a).

<table>
<thead>
<tr>
<th>Measurement area</th>
<th>3-D optical profilometer (160 nm/pixel)</th>
<th>AFM (40 nm/pixel)</th>
<th>SEM (0.9 nm/pixel)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.1 mm × 1.1 mm</td>
<td><img src="image1" alt="3-D optical profilometer image" /></td>
<td><img src="image2" alt="AFM image" /></td>
<td><img src="image3" alt="SEM image" /></td>
</tr>
<tr>
<td>30 μm × 30 μm</td>
<td><img src="image4" alt="3-D optical profilometer image" /></td>
<td><img src="image5" alt="AFM image" /></td>
<td><img src="image6" alt="SEM image" /></td>
</tr>
<tr>
<td>2 μm × 2 μm</td>
<td><img src="image7" alt="3-D optical profilometer image" /></td>
<td><img src="image8" alt="AFM image" /></td>
<td><img src="image9" alt="SEM image" /></td>
</tr>
</tbody>
</table>

**Figure 2.9** Surface characterization on the same interrogation area.
A broad area (e.g., 1.1 × 1.1 mm²) was characterized using a Taylor Hobson coherence correlation interferometry (CCI) 3-D optical profilometer at the Harvard Center for Nanoscale Systems (CNS). The optical profilometer enables non-contact surface measurements with sub-micron pixel resolutions (160 nm/pixel for the images in Figure 2.9). The surface topography of an area provides indication of the general surface morphology, and statistical parameters to quantify the 3-D surface roughness (e.g., $S_a$). In several previous studies, it was observed that not only micro-scale but also nano-scale surface features affect boiling [40, 45, 47]. Therefore, as second step, we characterized the nano-scale surface topography, as shown in Figure 2.9 (c), using a Bruker Dimension Icon Atomic Force Microscope (AFM) available at the Center for Materials Science and Engineering (CMSE) in MIT. A NANOSENSORS™ PPP-CONTPt AFM probe was used in tapping mode. The image in Figure 2.9 (c) correspond to the red square region in Figure 2.9 (a).

For comparison, we show in Figure 2.9 (b) a magnification of the image obtained with the optical profilometer (i.e., Figure 2.9 (a)) in the same exact position as Figure 2.9 (c). Note that the average surface roughness ($S_a$) and roughness ratio ($r$) measured with the two different techniques have similar values. However, the 3-D view in Figure 2.10 demonstrates that the profilometer data (187×187 pixel²) exhibit false peaks compared to AFM data (750×750 pixel²) due to lower measurement resolution. Therefore, the AFM was used for analysis of the local nano-scale topography (i.e., μm to nm scale).

![Figure 2.10](image-url)  
**Figure 2.10** Sub-micron surface topography on the same region measured by (a) 3-D optical profilometer (same data in Figure 2.9 (b)) and (b) AFM (same data in Figure 2.9 (c)).
In addition to surface morphology that may affect bubble triple contact line dynamics, another surface feature that significantly affects boiling is the presence of nucleation cavities. The size and shape of a nucleation cavity affects the bubble nucleation temperature and ultimately the departure diameter. In order to examine how potential nucleation cavities evolve as the surface is oxidized or coated, the shape of a certain cavity was imaged using a FEI Helios NanoLab™ 600 Dual Beam Scanning Electron Microscope (SEM) at MIT CMSE. Figure 2.9 (e) shows a SEM image of a ~2 μm size cavity with sub-nanometer precision, by zooming into the red square marked-region in Figure 2.9 (d). Note that Figures 2.9 (b), (c), and (d) were taken at the same area but using different techniques.

Surface wettability was estimated by the sessile droplet technique using a KSV CAM101 goniometer. The static contact angle of a ~1 μL sessile droplet was measured on the center of the boiling surface after each boiling test. Measurements were performed immediately after a sample was taken out from the flow loop in order to prevent measurement biases due to hydrophobic recovery, i.e., wettability of the surface decreases due to contamination, in air. Multiple contact angle measurements were conducted on the same sample to obtain an average contact angle value as well as the uncertainty range. Figure 2.11 shows an example of contact angle for multiple measurements. After each measurement, the surface was thoroughly cleaned by Isopropanol and DI water. Static contact angle reported in this study was calculated by averaging the saturated values, i.e., for the last five points in Figure 2.11.

Similarly, wicking was examined by the droplet spreading method discussed in Ref. [76]. The hemi-spreading process of a 1 μL water droplet was recorded.

![Figure 2.11 Static contact angle on the same surface and location for multiple measurements.](image)
2.5 Test Matrix

The test matrix of the current study includes multiple (and repeated) subcooled flow boiling experiments and surface characterizations, as reported in Table 2.1. The symbol “O” indicates completed surface characterization. The symbol “△” indicates surface characterization on several selected samples. The symbol “X” indicates that the measurement has not been made or does not apply to the selected surface. Note that sample 2 in each test is used to demonstrate the repeatability of the results. For the Cr-coated surface, tests on different substrates (Zircaloy-4 and FeCrAl) are used for this purpose, i.e., to provide reproducibility.

<table>
<thead>
<tr>
<th>Surface finish preparation method</th>
<th>Separate effect investigation</th>
<th>Sample type</th>
<th>Subcooled flow boiling CHF test</th>
<th>Surface characterization</th>
</tr>
</thead>
<tbody>
<tr>
<td>Random polishing</td>
<td>Substrate thickness</td>
<td>0.34 mm ~1.12 mm</td>
<td>Total 17 samples</td>
<td>△ (selected) X X △ X</td>
</tr>
<tr>
<td></td>
<td>Zircaloy-4 oxidation</td>
<td>Fresh</td>
<td>Sample 1</td>
<td>O O O O X</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Sample 2</td>
<td>O O O O X</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Pre-transition</td>
<td>Sample 1</td>
<td>O O O O X</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>Sample 2</td>
<td>O O O O X</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Transition</td>
<td>Sample 1</td>
<td>O O O O X</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>Sample 2</td>
<td>O O O O X</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Post-transition</td>
<td>Sample 1</td>
<td>O O O O O</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>Sample 2</td>
<td>O O O O O</td>
<td></td>
</tr>
<tr>
<td>Surface morphology</td>
<td>Streamwise</td>
<td>Sample 1</td>
<td>O X X X X</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>Sample 2</td>
<td>O X X X X</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Spanwise</td>
<td>Sample 1</td>
<td>X X X X X</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>Sample 2</td>
<td>X X X X X</td>
<td></td>
</tr>
<tr>
<td>Unidirectional polishing</td>
<td>Fresh</td>
<td>Zircaloy-4</td>
<td>Sample 1</td>
<td>O O O O X</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Sample 2</td>
<td>O X X O X</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Cr-coated</td>
<td>Zircaloy-4</td>
<td>Sample 1</td>
<td>O O O O X</td>
</tr>
<tr>
<td></td>
<td>FeCrAl</td>
<td>Fresh</td>
<td>Sample 1</td>
<td>O O O O X</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Sample 2</td>
<td>O X X O X</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Cr-coated</td>
<td>FeCrAl</td>
<td>Sample 1</td>
<td>O O O O X</td>
</tr>
</tbody>
</table>
Chapter 3

Post-processing Methodology

3.1 Infrared (IR) Post-processing

3.1.1 Heat Loss Study

A STAR-CCM+ model with simplified a heater-cartridge-fluid arrangement was developed to perform a heat loss study, as shown in Figure 3.1 (a). The fluid cross-section is $1 \times 3$ cm$^2$, i.e., the same dimension as the actual flow channel. An entrance region of 1-m length is used to ensure fully-developed flow conditions. There are three sections in the solid region: the active heating area of the Zircaloy-4 heater, the non-active heating area of the Zircaloy-4 heater, and the ceramic heater cartridge. Boundary and interface conditions are shown in Figure 3.1 (b). Constant power (i.e., constant areal heat generation rate or heat flux, $q''_a$) is applied to the active heating area and transferred to the non-active heating area of heater and cartridge by conduction. Heat is also transferred to air by convection (assuming a natural convection heat transfer coefficient of 10 W/m$^2$K [77]). The heat transfer between solid and fluid is computed by conjugate heat transfer simulations. The lateral surface is assumed adiabatic, and the thermal contact resistance of the air gap (<1 mm) between heater and cartridge is set to be 0.0005 m$^2$K/W, adopted from the thermal contact resistance between SS304 and ground air [78].

In steady-state conditions, the simulated total amount of heat transferred from the active surface to water, from the non-active surface to water, from the heater to the cartridge, and from the heater to air are shown in Figure 3.2. These results show that heat transferred from the active surface to water is dominant. Around 10% of heat is lost through the non-active surface. The heat transfer from the heater to the cartridge (0.71%) and to air (0.04%) is almost negligible, as it is, in total, less than 1% of the applied power. Note that this simulation was conducted for a single-phase fluid. The actual heat transferred from the active boiling area to water would be even higher, since the boiling heat transfer coefficient is much higher than single-phase forced convection heat transfer coefficient. This heat loss study confirms that the adiabatic boundary conditions used for the air side and the heater-cartridge interface in the following section is a reasonable assumption.
Figure 3.1 Heat loss study of the flow loop test section. (a) STAR-CCM+ model (b) Boundary and interface conditions.

Figure 3.2 Simulation results for heat loss study.
3.1.2 Inverse Conduction

A STAR-CCM+ computational model solving a 3-D inverse conduction problem was developed incorporating temperature-dependent properties (e.g., $\rho(T)$, $C_p(T)$, $k(T)$) of the metallic substrate (e.g., Zircaloy-4) to estimate the time-averaged temperature and heat flux on the boiling surface. The variables known a priori are the time-averaged temperature distribution obtained from the IR counts measured with the IR camera on the airside active heating area of a heater, $T_{back}(x, y)$, and the bulk temperature of the flow, $T_b$, measured by thermocouples, nominally 90 °C (i.e., 10 K subcooled flow boiling at atmospheric pressure).

The mesh (top view) of the heater model is shown in Figure 3.3 (a). The active area consists of the same number of cells as the experimental temperature data measured with the IR camera, $T_{back}(x, y)$, e.g., 87 × 87 cells for a 10 × 10 mm² area. Coarser mesh is used for the non-active area in order to reduce the computational costs. The volume mesh (i.e., through the heater thickness), is discretized with 10 μm thick cells. The boundary conditions are shown in Figure 3.3 (b). The heat loss study in Section 3.1.1 confirmed that the overall HTC at the airside, $h_a$, and the heater-cartridge interface, $h_l$, are small enough to consider these boundaries as adiabatic. The HTC between the non-active area and water, $h_{sp}$, is assigned as experimentally measured in single-phase forced convective conditions. The HTC on the active boiling area, $h_b$, is the unknown variable to evaluate iteratively.

![Figure 3.3](image)

**Figure 3.3** A heater model for 3-D inverse conduction. (a) 2-D mesh (b) Boundary conditions.
The schematic diagram of the inverse conduction model is shown in Figure 3.4.

![Schematic diagram of the 3-D inverse conduction model.](image)

**Figure 3.4** Schematic diagram of the 3-D inverse conduction model.

At the \(i\)-th iteration, the temperature distribution inside the substrate \(T(x, y, z, i)\) is estimated based on a guess value of \(h_b\), i.e., the boiling heat transfer coefficient. Given \(T_{\text{back}}(x, y)\), the heat flux to the substrate at the airside can be calculated by the discretized Fourier’s conduction law,

\[
q''_{\text{back}}(x, y, i) = k(T) \frac{T_{\text{back}}(x, y) - T(x, y, z_c, i)}{z_c} \tag{3.1}
\]

where \(z_c\) is the distance between the active heating area and the center of the adjacent cell, and \(k(T)\) is the substrate thermal conductivity for \(T(x, y, z_c, i)\). Then, \(h_b\) was updated iteratively until the solution converges and satisfies the criterion,

\[
\left( \frac{|q''_{\text{back}(i)} - V \cdot I/A_h|}{V \cdot I/A_h} \right) < 10^{-5} \tag{3.2}
\]

where \(V\) and \(I\) are the average measured voltage and current applied to the heater, respectively, and \(A_h\) is the active heating area. Therefore, \(V \cdot I/A_h\) is the experimentally applied heat flux, \(q''_{\text{applied}}\). Governing equations for this model consist of the heat balance and the Newton’s law of cooling, as follows:

\[
q''_{\text{applied}} A_h = q''_{\text{boiling}} A_{\text{boiling}} + q''_{\text{sp}} A_{\text{sp}} \tag{3.3}
\]

\[
q''_{\text{boiling}} = h_b (T_{\text{boiling}} - T_b) \tag{3.4}
\]

\[
q''_{\text{sp}} = h_{sp} (T_{sp} - T_b) \tag{3.5}
\]

From Eq. (3.3) to (3.5), the boiling HTC, \(h_b\), can be evaluated as below:

\[
h_b = \frac{q''_{\text{applied}} A_h - h_{sp} (T_{sp} - T_b) A_{sp}}{(T_{\text{boiling}} - T_b) A_{\text{boiling}}} \tag{3.6}
\]
Finally, the formula to update \( h_b \) implemented in STAR-CCM+ is:

\[
\begin{align*}
    h_b^{i+1} &= \frac{Q_{\text{applied}} - \int_A h_sp(T_{sp} - T_b)}{\int_A (T_{boiling} - T_b)} \\
    \text{(3.7)}
\end{align*}
\]

Once the iterative process has converged, the heat flux distribution to water, \( q_w''(x,y) \), can be calculated as:

\[
q_w''(x,y) = k(T)\frac{T(x,y,z_{cr}) - T_{boiling}(x,y)}{z_{cr}}
\]

where \( z_{cr} \) is the distance between the active boiling area and the center of the adjacent cell, \( k(T) \) is the substrate thermal conductivity for \( T(x,y,z_{cr},i) \), and \( T_{boiling}(x,y) \) is the converged wall temperature distribution. The boiling curve can be obtained by space-averaging the temperature and heat flux distributions within the uniform central region of the boiling surface, \( T_{boiling} \) and \( \overline{q_w''} \), respectively.

A simpler 1-D post-processing script was also developed in MATLAB to facilitate faster processing of boiling curves. In the 1-D inverse conduction problem, a single temperature value on the airside active heating area, \( \overline{T_{back}} \), is obtained by averaging the temperature distribution, \( T_{back}(x,y) \), over the central region of the heater which is at uniform temperature, as shown in Figure 3.5. With the known heat flux value, \( q_{\text{applied}}'' \), assuming no heat loss within this region, temperature at the boiling surface can be calculated by the 1-D conduction equation, as follows:

\[
\overline{T}(z + \Delta z) = \overline{T}(z) - q_{\text{applied}}'' \frac{\Delta z}{k(\overline{T}(z))}
\]

where \( \Delta z \) is a discretized step size.

![Figure 3.5 Schematic diagram of the 1-D inverse conduction model.](image)
The thermal conductivity of a Zircaloy-4 specimen was calculated by the following equation:

\[
k(T) = \alpha(T) \cdot \rho(T) \cdot c_p(T)
\]  
(3.10)

where \( \alpha \) is the thermal diffusivity, \( \rho \) is the density, and \( c_p \) is the specific heat capacity. The thermal diffusivity of a specimen was measured using a Netzsch NanoFlash® Laser Flash Analysis (LFA) 447 at the MIT Nuclear Reactor Laboratory (NRL). Correlations for the temperature dependent density and heat capacity of Zircaloy-4 were taken from Ref. in [79] as listed below,

\[
c_p(T) = 255.66 + 0.1024T \ [J/kgK] \ (273 K < T < 1100 K)
\]  
(3.11)

with 2~3% uncertainty (i.e., \( \pm 10 \) J/kg-K).

\[
\rho(T) = 6595.2 - 0.1477T \ [kg/m^3] \ (T < 1083 K)
\]  
(3.12)

with 0.67% uncertainty.

Calculated thermal conductivities for Zircaloy-4 obtained with three independent thermal diffusivity measurements are shown in Figure 3.6. A quadratic fitting gives,

\[
k(T) = 1.5173 \times 10^{-5}T^2 - 8.7 \times 10^{-3}T + 15.4788 \ [W/mK]
\]  
(3.13)

with \( \pm 0.23 \) W/m-K uncertainty range.

Eq. (3.13) was implemented in the inverse conduction solvers for Zircaloy-4.

![Figure 3.6 Calculated thermal conductivity for Zircaloy-4.](image)

For the commercial Kanthal APMT FeCrAl alloy, Eq. (3.14) (from [80]) was implemented in the inverse conduction solvers.
\begin{equation}
k(T) = -7.223 \times 10^{-7} T^2 + 1.5628 \times 10^{-2} T + 6.569 \text{ [W/mK]} \tag{3.14}
\end{equation}

with \(\pm 0.74\) W/m-K uncertainty range.

Temperature drops due to low-conductivity layer (e.g., \(\text{Al}_2\text{O}_3\) insulation layer, Zircaloy-4 oxide layer) was taking into account as follows,

\begin{equation}
\Delta T_o = q''_{\text{applied}} \cdot \frac{\delta_o}{k_o} \tag{3.15}
\end{equation}

where \(\delta_o\) is the thickness of a layer and \(k_o\) is the thermal conductivity of low-conductive material. A constant value of 2.2 W/m-K was used for \(\text{Al}_2\text{O}_3\) assuming that the property change due to the temperature gradient within the \(~4\) \(\mu\)m of the insulation layer is negligible. Temperature dependent thermal conductivity of \(\text{ZrO}_2\) was adopted from [81] and calibrated to estimate the single-phase HTC of the oxidized Zircaloy-4 to be the same as the fresh Zircaloy-4:

\begin{equation}
k_o(T) = 3.62 \times 10^{-4} T + 1.67 \text{ [W/mK]} \tag{3.16}
\end{equation}

The thermal conductivity in Eq. (3.16) ranges between 1.8~1.85 W/m-k within our interested temperature range (e.g., from the single-phase to CHF). These values agree rather well with the thermal conductivity of \(\text{ZrO}_2\), 1.6~2.2 W/m-K, used in the modeling of fuel performance, e.g., in Refs. [81-83].
3.1.3 Uncertainty Analysis

The uncertainty of the calculated wall superheat was analyzed by the error propagation method. Based on 1-D conduction within a substrate, temperature at the boiling surface, $T_{wall}$, is expressed as follows:

$$T_{wall} = T_{back} - q''_{applied} \cdot \frac{\delta}{k}$$

(3.17)

where $T_{back}$ is the temperature at the airside surface, $q''_{applied}$ is the applied heat flux, $\delta$ is the substrate thickness, and $k$ is the thermal conductivity of the substrate. The uncertainty of $T_{wall}$, $\Delta T_{wall}$, is given by

$$\Delta T_{wall}^2 = \left(\frac{\partial T_{wall}}{\partial T_{back}}\right)^2 \Delta T_{back}^2 + \left(\frac{\partial T_{wall}}{\partial q''_{applied}}\right)^2 \Delta q''_{applied}^2 + \left(\frac{\partial T_{wall}}{\partial \delta}\right)^2 \Delta \delta^2 + \left(\frac{\partial T_{wall}}{\partial k}\right)^2 \Delta k^2$$

$$= \Delta T_{back}^2 + \left(\frac{\delta}{k}\right)^2 \Delta q''_{applied}^2 + \left(\frac{q''_{applied}}{k}\right)^2 \Delta \delta^2 + \left(\frac{q''_{applied} \cdot \delta}{k^2}\right)^2 \Delta k^2$$

(3.18)

$q''_{applied}$ is in the order of 2~3 MW/m$^2$ near CHF in the operating conditions of this work, $k$ is in the order of 10~20 W/m-K within the interested temperature range, and $\delta$ is in the order of 0.5~1 mm for metallic heaters used in the present work. In order to evaluate the maximum uncertainty value for general experimental conditions, Eq. (3.18) near CHF is approximated as below:

$$\Delta T_{wall}^2 = \Delta T_{back}^2 + (10^{-5})^2 \Delta q''_{applied}^2 + (10^5)^2 \Delta \delta^2 + (5)^2 \Delta k^2$$

(3.19)

The uncertainty of each variable is analyzed hereafter.

- $\Delta T_{back}$

$T_{back}$ is obtained by averaging 2-D temperature distribution at the airside, $T_{back}(x, y)$, calibrated from IR counts. Since calibration error is very small, $\Delta T_{back}$ mostly depends on the spatial variation of the 2-D temperature distribution. Typical spatial deviation of the 2-D temperature distribution within the uniform central region is 5~10 °C.

- $\Delta q''_{applied}$

$q''_{applied}$ is calculated by $V \cdot I/A_h$. Its propagated uncertainty is formalized below:

$$\Delta q''_{applied}^2 = \left(\frac{I}{A_h}\right)^2 \Delta V^2 + \left(\frac{V}{A_h}\right)^2 \Delta I^2 + \left(\frac{V I}{A_h^2}\right)^2 \Delta A_h^2$$

(3.20)
The order of the applied voltage \( (V) \) is 30 V near CHF with the measurement uncertainty \( (\Delta V) \) of 0.001 V from high-speed data acquisition system (HSDAS) [84]. The order of the current \( (I) \) to a heater is 10 A near CHF with the measurement uncertainty \( (\Delta I) \) of 0.02 A from HSDAS and voltage dividers [84]. The order of the active heating area \( (A_n) \) is \( 10 \times 10 \text{ mm}^2 \) with the measurement uncertainty \( (\Delta A_n) \) of \( 0.02 \times 0.02 \text{ mm}^2 \) using a caliper. Overall, the uncertainty of the applied heat flux \( (\Delta q''_{\text{applied}}) \) is \( \sim 10 \text{ kW/m}^2 \). Therefore, the effective temperature deviation due to the second term in eq. (3.19) is less than 1 °C, which is practically negligible compared to \( \Delta T_{\text{back}} \).

- \( \Delta \delta \)

\( \delta \) is measured by a caliper with a precision of 0.01 mm. A heater may have thickness variation due to non-uniform polishing. Typical measurement uncertainty of a substrate thickness \( (\Delta \delta) \) is 0.01~0.015 mm. Therefore, the effective temperature deviation due to the third term in eq. (3.19) is 1~3 °C, which is relatively smaller than \( \Delta T_{\text{back}} \).

- \( \Delta k \)

\( \Delta k \) is estimated to be 1~2 W/m-K based on the uncertainties of eq. (3.13) and (3.14). Therefore, the effective temperature deviation due to the forth term in eq. (3.19) is 5~8 °C.

Based on the above analysis, the uncertainties associated with the 2-D temperature distribution \( (\Delta T_{\text{back}}) \) and the thermal conductivity \( (\Delta k) \) are the dominant ones to determine \( \Delta T_{\text{wall}} \) in Eq. (3.19). The standard deviation of the 2-D temperature distribution is different in every test, whereas the same correlation of the thermal conductivity is used for the same substrate material. Therefore, only the spatial uncertainty range of 2-D wall superheat distribution \( (T_{\text{wall}}(x, y)) \) was shown within the boiling curves in this study.
3.2 High-speed Video (HSV) Post-processing

3.2.1 Bubble Image Segmentation

For bubble image segmentation, a deep-learning tool, U-Net [85], was integrated in the HSV post-processing protocol. U-net was originally developed to detect cells in biological images. Since cells and bubbles have common features (e.g., they are mostly round but may evolve to an irregular shape, or have non-uniform contrast and background texture), U-net can be effectively trained with a small set of ground-truth bubble images to recognize and segment boiling bubbles from HSV images with high accuracy, no matter the bubble size (e.g., large or small) or shape (e.g., regular or irregular) and the image contrast or the background texture. The contrast and appearance of boiling bubbles vary depending on the position and reflection of background lights, in conjunction with the surface texture, especially for a rough surface. Figure 3.7 (a) shows examples of high-speed imaging in subcooled flow boiling on different metallic heaters.

![Figure 3.7](image)

**Figure 3.7** High-speed imaging and bubble segmentation on various surface conditions in subcooled flow boiling. (a) Raw HSV data (b) Bubble segmentation by U-net (c) Binary bubble mask.
Bubbles have diverse size and contrast and the background (i.e., boiling surface) has different contrast, texture and morphology, due to the different polishing method or different surface material compositions (e.g., unoxidized or oxidized surfaces). In these cases, typical threshold-based image processing [60-61, 73, 75] or machine vision [8, 74] techniques are not well applicable, because those techniques require a certain image quality or bubble illumination. On the contrary, U-Net is highly adaptable, as shown in Figure 3.7 (b). Particularly, the segmentation process which separates adjacent objects is more adaptive than typical morphological transformation used in image-processing (e.g., a watershed).

For the HSV images post-processing, U-Net was first pre-trained with 6 manually annotated bubble images at different heat flux levels for 10000 iterations, and the pre-trained model was validated with 2 manually annotated bubble images every 1000 iterations. If HSV imaging of a new experiment has different appearance or quality, the U-net bubble segmentation model can be finetuned for each experiment with a small set of manually annotated images, typically less than 10, for 5000~20000 iterations.

Figure 3.7 (c) shows binary images, the so called “bubble mask,” which were used in the following post-processing steps.
3.2.2 Bubble Tracking

The segmented bubble masks were classified into individual and coalesced patches. Individual patches were tracked along their trajectory until they either coalesced or completely condensed. Bubble tracking can be done by different techniques, such as particle tracking velocimetry [75] or template matching [86-87]. In this study, since high-resolution HSV was recorded at 10000 fps, successive bubble patches are highly overlapped, as shown in Figure 3.8.

![Overlapped bubble masks in two successive frames](image)

**Figure 3.8** Overlapped bubble masks in two successive frames (sampled from Cr-coated FeCrAl at 1.2 MW/m²).

Therefore, matching candidates between the sequential bubble patches can be easily specified as the ones that have the overlapped region. A function, $E_{ij}$, was defined as Eq. (3.21) to set criteria to determine whether a bubble patch in the current frame, $f$, is a coalesced patch or an individual patch (the rationale is explained hereafter), and if it is individual, decide which bubble patch in the following frame, $f+1$, is the same as the current one, i.e., find the best matching pair. $E_{ij}$ indicates differences in location and shape of the sequential overlapping bubble patches, $i$ and $j$:

$$E_{ij} \equiv w_{xy} \sqrt{(x_i - x_j)^2 + (y_i - y_j)^2} + w_{ab} \sqrt{(a_i - a_j)^2 + (b_i - b_j)^2}$$  \hspace{1cm} (3.21)

where $(x, y)$ is the coordinate of the center of mass of a bubble patch, and $a$ is the major axis length and $b$ is the minor axis length of equivalent ellipse of a bubble patch, respectively. When there are multiple patches in the following frame ($j=1,2,3, ..$) that overlapped with the current
frame patch, \( i \), as illustrated in Figure 3.9 (a), the best matching object is determined as the one that gives the minimum value of \( E_{i,j} \), i.e., it is given by the combination with the minimum displacement and shape change. When there are multiple patches in the current frame \((i=1,2,3,..)\) that overlapped with a patch, \( j \), in the following frame, as illustrated in Figure 3.9 (b), the object \( j \) is classified as a coalesced patch if the minimum difference of \( E_{i,j} \) \((i=1,2,3,..)\) is small, e.g.,

\[
\min \left( \frac{E_{2j}}{E_{1j}} \right) < \alpha, \quad (E_{1j} < E_{2j})
\]

where \( \alpha \) is an user-defined parameter. When bubbles with similar size are coalescing, \( \alpha \) should be small, close to 1. When coalescing bubbles have rather different sizes, \( \alpha \) would be larger. In this case, bubble coalescence does not have much impact, and the coalesced patch generally follows the motion of a larger bubble from the previous frame. In this study, \( \alpha \) is set to be 1.5. A bubble patch matched with the previously defined-coalesced patch is also considered as a coalesced patch.

**Figure 3.9** Conceptual examples for non-one-to-one matching cases. (a) Multiple matching candidates at the following frame (b) Bubble coalescence.
### 3.2.3 Bubble Life-cycle Stage Identification

Identifying bubble life-cycle stages, e.g., nucleation, growth, sliding, and condensation, is important to extracting relevant boiling parameters, which are then used in the heat flux partitioning models. A life-cycle stage for an individual bubble patch was identified at each frame. Motion between successive bubble masks is very small due to the high temporal resolution of the HSV, i.e., 10000 fps. Therefore, a global optical flow algorithm [85], which is beneficial in small motion estimation, was used to compute the displacement field, \( \vec{d} = (d_x, d_y) \), and its divergence map, \( \nabla \cdot \vec{d} \), between successive bubble masks (see Figure 3.10 (b)). The averaged divergence and y-direction (i.e., parallel to flow) displacement within each bubble region (Figure 3.10 (c) and (d)) were used to determine the life-cycle stage.

**Figure 3.10** Bubble motion estimation using an optical flow algorithm. (a) Snapshot of HSV (b) Computed displacement vectors (arrow) and divergence map (color) (c) Bubble region-averaged divergence (d) Bubble region-averaged y-direction displacement.
Qualitative trends of divergence and y-direction displacement during a bubble life cycle are shown in Figure 3.11. When a bubble nucleates and grows, the divergence is strongly positive. Instead, when a bubble departs from the surface and condenses, the divergence becomes negative. Therefore, negative divergence was considered as an indicator of the moment when a bubble leaves the surface, and no longer contributes to wall boiling heat transfer. A conventional method to determine bubble sliding moment is to check if the center of mass of a bubble deviates from the nucleation site. However, for a large bubble, even the center of mass point deviates from the nucleation site, the bubble may just roll while growing. In order to discriminate bubble rolling motion from actual sliding, another criterion was set: y-component displacement should be strongly negative when a bubble starts to slide.

**Figure 3.11** Qualitative trends of divergence and y-direction displacement, $v$, during the bubble life-cycle.
The performance of the overall HSV post-processing approach (i.e., bubble segmentation, tracking, and life-cycle identification) is qualitatively demonstrated in Figure 3.12 on images obtained using a phase-detection technique on a sapphire-ITO heater [55], which tracks dry area, microlayer, and vapor phases on the boiling surface. Dry spot inside a bubble shows clear evidence of a bubble attached to the surface. The quantitative assessment of the bubble life-cycle stages identification is done by extracting corresponding parameters in the following section. The trajectory of a bubble patch is tracked (green line) from its nucleation site until the bubble completely condenses or coalesces. Similarly, the post-processing approach is well applicable to HSV imaging on a metallic heater, as shown in Figure 3.13.

**Figure 3.12** Demonstration of the performance of the HSV post-processing algorithm to a phase-detection image for smooth ITO. Raw data (left) and post-processed result (right).

**Figure 3.13** Application of the HSV post-processing algorithm to a metallic heater. Raw data (left) and post-processed result (right).
3.2.4 Validation using Sapphire-Zr Rough Heater

The post-processing approach discussed in the previous section was validated by comparing the measured boiling parameters with those measured during the same exact experiment by infrared thermometry using a special heater [54]. The heater enables both infrared and shadowgraph investigations on a roughened metallic surface by using a sapphire substrate, as shown in Figure 3.14. HSV images were not recorded at high heat flux because in those conditions all bubbles merge creating a vapor patch that obstructs optical access to the surface.

\[\text{Figure 3.14 Sapphire-Zr rough heater design.}\]

For the HSV, nucleation sites were identified by circular object detection on the activity map obtained by time-averaging nucleation bubble patches, as shown in Figure 3.15. For the IR, nucleation sites were picked manually, by frame-by-frame observations. Number and location of nucleation sites obtained by HSV and IR post-processing are compared in Figure 3.16 (a) and (d), respectively. The measurements obtained by the two diagnostics are in good agreement, both in
terms of nucleation site density and the local position of the nucleation sites. In the post-processing of the HSV images, nucleation events were recorded for each nucleation site to calculate the bubble period. The averaged nucleation frequency over all nucleation sites is compared in Figure 3.16 (b). In the IR investigation, the effective bubble footprint area was sampled based on heat flux level. The average radius of the IR bubble footprint mask and the average radius of HSV bubble mask segmented by U-net are compared in Figure 3.16 (c), and shows good agreement about the characteristic bubble size.

The comparison in Figure 3.16 demonstrates that the HSV post-processing approach provides convincing quantification ability on near-wall bubble dynamics up to intermediate heat flux.

![Identification of nucleation sites. (a) Nucleation activity map (b) Detected nucleation sites.](image)

**Figure 3.15** Identification of nucleation sites. (a) Nucleation activity map (b) Detected nucleation sites.
Figure 3.16 Quantitative comparison of the performance of IR and HSV post-processing techniques. (a) Nucleation site density (b) Nucleation frequency (c) Average radius of bubble area (d) Location of nucleation sites.
Chapter 4

Heat Flux Partitioning (HFP) Modeling

4.1 HFP Modeling for Subcooled Flow Boiling

Heat flux partitioning (HFP) is a very popular approach to model boiling heat transfer. One of the most popular model is the so-called RPI model [89], according to which the total heat flux is given by the sum of three contributions: evaporation, quenching, and single-phase convective heat flux. In flow boiling conditions, heat is additionally transferred by sliding bubbles [90-94].

The evaporation term is often calculated through the volume of the vapor bubble, which is created by the evaporation of the microlayer and the superheated layer around the bubble, as shown in Figure 4.1 (c). However, the energy transferred through the superheated layer is already taken into account in the quenching term, as shown in Figure 4.1 (e) (a) (b). Therefore, in recent studies, the evaporation term is often expressed as the microlayer evaporation term only [53-54, 92].

![Figure 4.1 Bubble life cycle. (a) Formation of vapor embryo (b) Initial bubble growth due to the evaporation of superheated boundary layer and single-phase convection (c) Bubble growth due to the evaporation of microlayer and superheated boundary layer (d) Bubble departure (e) Quenching.](image)

HFP models [89-91, 93, 95-97] use averaged boiling parameters (e.g., departure frequency and wait time, and microlayer outer radius) and nucleation site density as input parameters, but do not
consider the temporal aspects of the bubble interaction process. Practically, they assume that bubbles nucleate at the same time (i.e., they are in-phase) and with the same size from all nucleation sites, as illustrated in Figure 4.2 (a). However, these nucleation sites are not all sustaining bubbles at the same time, e.g., because the growth of a bubble from one nucleation site may obstruct other potentially active sites. In order to consider the fraction of nucleation sites that can simultaneously sustain bubbles, i.e., sites that are concurrently activated, recent studies applied a complete spatial randomness (CSR) approximation to evaluate the effective nucleation site density [92] or a stochastic modeling of the bubble interaction process [54], as shown in Figure 4.2 (b).

In this study, we can quantify boiling parameters at each nucleation site using the HSV images processing technique discussed in Chapter 3. Then, each partitioned heat flux term can be calculated for each nucleation site, and summed over the whole boiling surface (i.e., summed over all the nucleation sites). In this way, we account for the fact that bubbles nucleate at different times and with different size depending on the nucleation site, as shown in Figure 4.2 (c).

![Figure 4.2](image)

**Figure 4.2** Sketch of active nucleate bubble scenarios postulated in different HFP modelling (a) Using averaged quantities for all nucleation sites (b) Modeling of active nucleation sites (c) Using distinct quantities for each nucleation site.

When we assume that thermal conduction limits the bubble growth, microlayer thickness can be estimated as a linear relationship with the bubble radius:

\[
\delta(r) = C \cdot r
\]

(4.1)

where \(C\) is a parameter expressed as \(\sqrt{\frac{t_b v_l}{2r_b}}\), where \(t_b\) is the time for a bubble to grow to the radius \(r_b\), and \(v_l\) is the kinematic viscosity of the liquid [54]. During the initial stage of its growth, the bubble grows symmetrically, as shown in Figure 4.3 (a).
The volume of microlayer of radius $R$ is given by

\[ V_R = 2\pi \int_0^R \delta(r) \, r \, dr \]
\[ = \frac{2}{3} \pi CR^3 \]  

(4.2)

However, during sliding, bubble is elongated in the flow direction and the microlayer grows asymmetrically, as shown in Figure 4.3 (b). The volume of microlayer can be expressed with the asymmetric radial lengths, $R_{min}$ and $R_{max}$, their corresponding thickness, $\delta_{R_{min}}$ and $\delta_{R_{max}}$, the major axis length of the equivalent ellipse, $a$, and the minor axis length of the equivalent ellipse, $b$. Note that, $2a = R_{min} + R_{max}$. With these definitions, and assuming the microlayer thickness is proportional to the distance from the nucleation site, the volume of the asymmetric microlayer can be geometrically derived as follows:

\[ V_{ab} = \pi ab \left( \frac{\delta_{R_{min}} + \delta_{R_{max}}}{2} - \frac{1}{6} \frac{R_{max} \delta_{R_{min}} + R_{min} \delta_{R_{max}}}{a} \right) \]  

(4.3)
Given a constant, linear microlayer growth rate, as shown in Eq. (4.1), and plugging in \( \delta_{R_{\text{min}}} = C \cdot R_{\text{min}} \) and \( \delta_{R_{\text{max}}} = C \cdot R_{\text{max}} \) into Eq. (4.3), we obtain

\[
V_{ab} = \pi ab \left( \frac{C(R_{\text{min}} + R_{\text{max}})}{2} - \frac{1}{3} \frac{C^2 R_{\text{min}} R_{\text{max}}}{a} \right), R_{\text{min}} \in [0, a], R_{\text{max}} \in [a, 2a]
\] (4.4)

When \( R_{\text{min}} = R_{\text{max}} = a \), Eq. (4.4) becomes,

\[
V_{ab} = \frac{2}{3} \pi C b a^2
\] (4.5)

When \( R_{\text{min}} = 0, R_{\text{max}} = 2a \), Eq. (4.4) becomes,

\[
V_{ab} = \pi C b a^2
\] (4.6)

Based on Eq. (4.5) and Eq. (4.6), the maximum modeling uncertainty is ~33%. In this study, Eq. (4.2) was used to calculate the symmetric microlayer volume, and Eq. (4.5) was used to calculate the asymmetric microlayer volume.

In this study, heat transfer mechanisms by a “static” bubble and a “moving” bubble are assessed separately to discriminate the effect of flow. The proposed HFP model is summarized in Eqs. (4.7) throughout (4.11):

- Static evaporation:
  \[
  q_{e''} = \sum_{i=1}^{N} \left( f_i \frac{V_{\mu_{l,i}}}{A_h} \right) \rho_l h_{fg} \text{ with } V_{\mu_{l,i}} = \frac{\sqrt{r_{g,i}^2 + r_{l,i}^2}}{3} \left( \pi r^2_{a_{l,i}} \right)
  \] (4.7)

This term indicates microlayer evaporation at the initial growth stage when a bubble grows around its nucleation site.

- Sliding bubble evaporation:
  \[
  q_{s,e''} = \sum_{i=1}^{N} \left( f_i \frac{V_{\mu_{l,\text{lift},i}} - V_{\mu_{l,i}}}{A_h} \right) \rho_l h_{fg} \text{ with } V_{\mu_{l,\text{lift},i}} = \frac{\sqrt{(t_{g,i} + t_{s,i})^2}}{3} \left( \pi a_{l,\text{lift},i} b_{\text{lift},i} \right)
  \] (4.8)

This term calculates the volume addition due to microlayer evaporation during the sliding of the bubble. The symmetric microlayer volume at the static stage is subtracted from the asymmetric
microlayer volume when a bubble lifts-off the surface.

- Static quenching:
  \[
  q_q'' = \sum_{i=1}^{N} \left( f_i \frac{\pi (r_{d,i}^2 + a_{lift,i} b_{lift,i})}{2} \sqrt{t_{w,i}/A_h} \right) K \epsilon_i \sqrt{\pi \Delta T}
  \]  
  (4.9)

This term includes averaged static quenching when a bubble departs from its nucleation site and a bubble lifts-off from the surface.

- Sliding conduction:
  \[
  q_{sc}'' = \sum_{i=1}^{N} \left( f_i (r_{d,i} + b_{lift,i}) \ l_{s,i} \sqrt{t_{s,i}/A_h} \right) K \epsilon_i \sqrt{\pi \Delta T}
  \]  
  (4.10)

This term indicates quenching in the wake of a bubble sliding.

- Forced convection:
  \[
  q_{fc}'' = \max \left[ 0, \left( 1 - \sum_{i=1}^{N} \left( (r_{d,i} + b_{lift,i}) l_{s,i} + \frac{\pi (r_{d,i}^2 + a_{lift,i} b_{lift,i})}{2} \right)/A_h \right) h_{fc} \Delta T \right]
  \]  
  (4.11)

where \( N \) is total number of nucleation sites, \( A_h \) is active heating area, \( r_{d,i} \) is averaged departure radius for \( i \)-th nucleation site, \( a_{lift,i} \) is averaged lift-off maximum microlayer axis for \( i \)-th nucleation site, \( b_{lift,i} \) is averaged lift-off minimum microlayer axis for \( i \)-th nucleation site, \( f_i \) is averaged nucleation frequency for \( i \)-th nucleation site, \( t_{w,i} \) is averaged wait time for \( i \)-th nucleation site, \( t_{g,i} \) is averaged growth time for \( i \)-th nucleation site, \( t_{s,i} \) is averaged sliding time for \( i \)-th nucleation site, and \( l_{s,i} \) is averaged sliding length for \( i \)-th nucleation site.

Since shadowgraph only visualizes vapor bubble, we used the HSV post-processed vapor bubble radius instead of the microlayer footprint radius in this study. The proposed HFP model is compared with the previous works in Table 4.1. In the current model, the quantified parameters at each nucleation site directly participate in calculating partitioned heat fluxes without additional modeling of interactions between nucleation sites. Also, asymmetric microlayer growth was considered. The proposed modeling strategies are evaluated in Section 5.3.3.
Table 4.1. Comparison of HFP models (continued)

<table>
<thead>
<tr>
<th>Microlayer evaporation</th>
<th>Static, $q_e''$</th>
<th>Sliding, $q_{s,e}''$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sateesh et al. (2005) [95]</td>
<td>$(N''f V_v) \rho_v h_{fg}$</td>
<td>$(N_s'' f (V_{lift} - V_v)) \rho_v h_{fg}$</td>
</tr>
<tr>
<td></td>
<td>$V_v = \frac{4\pi r_d^3}{3}$</td>
<td>$V_{lift} = \frac{4\pi r_{lift}^3}{3}$</td>
</tr>
<tr>
<td></td>
<td>$N_a'' = N''f t_{f g}$</td>
<td>$N_s'' = N''f t_{s}$</td>
</tr>
<tr>
<td>Gilman &amp; Baglietto (2017) [92]</td>
<td>$(N_a'' f V_{\mu l}) \rho_l h_{fg}$</td>
<td>$(N_a'' f D_{avg} l_s^2 \delta_{\mu l}) \rho_l h_{fg}$</td>
</tr>
<tr>
<td></td>
<td>$V_{\mu l} = \frac{2\pi (r_d^2)}{3} \delta_{\mu l}$</td>
<td>$D_{avg} = r_d + r_{lift}$</td>
</tr>
<tr>
<td></td>
<td>$N_a'' = N''f t_{f g}$</td>
<td>$N''f t_{s}$</td>
</tr>
<tr>
<td></td>
<td>$N_a'' = N''(N_{f g}^l + t_{s})\rho_l h_{fg}$</td>
<td>$N''f t_{f g}$</td>
</tr>
<tr>
<td>Su et al. (2020) [54]</td>
<td>$(N_b'' f V_{\mu l}) \rho_l h_{fg}$</td>
<td>$(N_a'' f D_{avg} l_s^2 \delta_{\mu l}) \rho_l h_{fg}$</td>
</tr>
<tr>
<td></td>
<td>$V_{\mu l} = \sqrt{\frac{t_{g,\mu l}^2}{3}} (\pi r_{\mu l}^2)$</td>
<td>$D_{avg} = r_d + r_{lift}$</td>
</tr>
<tr>
<td></td>
<td>$N_b'' = N''f t_{f g}$</td>
<td>$N''f t_{s}$</td>
</tr>
<tr>
<td></td>
<td>$N_b'' = N''(N_{f g}^l + t_{s})\rho_l h_{fg}$</td>
<td>$N''f t_{f g}$</td>
</tr>
<tr>
<td>This work</td>
<td>$\sum_{i=1}^{N} (f_i V_{\mu l,i}/A_h) \rho_l h_{fg}$</td>
<td>$\sum_{i=1}^{N} (f_i (V_{\mu l,i,lift,i} - V_{\mu l,i})/A_h) \rho_l h_{fg}$</td>
</tr>
<tr>
<td></td>
<td>$V_{\mu l,i} = \sqrt{\frac{t_{g,i}^2}{3}} (\pi r_{d,i}^2)$</td>
<td>$V_{\mu l,lift,i} = \sqrt{\frac{(t_{g,i}^2 + t_{s,i}^2)^{\nu_i}}{3}} (\pi a_{lift,i} b_{lift,i})$</td>
</tr>
</tbody>
</table>
Table 4.1. Comparison of HFP models (continued)*

<table>
<thead>
<tr>
<th>Model</th>
<th>Transient conduction</th>
<th>Forc ed convection, $q''_{fc}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Static, $q''_q$</td>
<td>$(N'' f \pi r_d^2 \sqrt{t_w}) \varepsilon_l \frac{2}{\sqrt{\pi}} \Delta T$</td>
<td>$N''<em>{s} f D</em>{avg} l_s \sqrt{t_w} \varepsilon_l \frac{2}{\sqrt{\pi}} \Delta T$</td>
</tr>
<tr>
<td>Sliding, $q''_{sc}$</td>
<td>$(N''<em>s f D</em>{avg} l_s \sqrt{t_w}) \varepsilon_l \frac{2}{\sqrt{\pi}} \Delta T$</td>
<td></td>
</tr>
<tr>
<td>Sateesh et al. (2005) [95]</td>
<td>$(N''<em>a f V_q) \rho_n c</em>{ph} \Delta T_h$</td>
<td>$(N''<em>{a*} f D</em>{avg} l_s^* \sqrt{t^*}) \varepsilon_l \frac{2}{\sqrt{\pi}} \Delta T$</td>
</tr>
<tr>
<td></td>
<td>$V_q = \frac{2}{3} (r_{dry})^2$</td>
<td>$N''<em>{a*} = \frac{1}{1+1/l_s} N''</em>{a*}$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$t^* = \frac{1}{\pi} \left( \frac{\varepsilon_l}{h_{fc}} \right)^2$</td>
</tr>
<tr>
<td>Gilman &amp; Baglietto (2017) [92]</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Su et al. (2020) [54]**</td>
<td>$(N''<em>{b*} f \pi r</em>{\mu l}^2 \sqrt{t_w}) \varepsilon_l \sqrt{\pi} \Delta T$</td>
<td></td>
</tr>
<tr>
<td>This work**</td>
<td>$\sum_{i=1}^{N} \left( f_i \frac{\pi (r_{d,l}^2 + a_{lift,l} b_{lift,l})}{2} \sqrt{t_{w,i}} / A_h \right) \varepsilon_l \sqrt{\pi} \Delta T$</td>
<td>$\sum_{i=1}^{N} \left( f_i \left( r_{d,i} + b_{lift,i} \right) l_{s,i} \sqrt{t_{s,i}} / A_h \right) \varepsilon_l \sqrt{\pi} \Delta T$</td>
</tr>
</tbody>
</table>

* Set K=1.

** assumed $t_q \cong t_w$.

Table 4.1. Comparison of HFP models*

<table>
<thead>
<tr>
<th>Model</th>
<th>Two-phase area: $A_{2\theta}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Static, $q''_{v}$</td>
<td>$N'' A_v + N''<em>{s} f t_w (D</em>{avg} l_s + A_{avg})$</td>
</tr>
<tr>
<td>Sliding, $q''_{vt}$</td>
<td>$N''<em>{a*} f t^* D</em>{avg} l_s^*$</td>
</tr>
<tr>
<td>Sateesh et al. (2005) [95]</td>
<td>$N'' A_{\mu l}$</td>
</tr>
<tr>
<td>Gilman &amp; Baglietto (2017) [92]</td>
<td>$N''<em>{b*} A</em>{\mu l}$</td>
</tr>
<tr>
<td>Su et al. (2020) [54]</td>
<td>$\sum_{i=1}^{N} \left( r_{d,i} + b_{lift,i} \right) l_{s,i} \sqrt{t_{s,i}} / A_h$</td>
</tr>
<tr>
<td>This work</td>
<td>$\sum_{i=1}^{N} \left( \frac{\pi (r_{d,i}^2 + a_{lift,i} b_{lift,i})}{2} \right) / A_h$</td>
</tr>
</tbody>
</table>

* Set K=1.
4.2 Modeling of Individual Bubble Size Distribution at High Heat Flux

Existing HFP models use as input boiling parameters (e.g., bubble departure diameter and bubble lift-off diameter) for discrete, non-interacting bubbles. However, at high heat fluxes, bubbles coalesce, and the bubble interaction affects these parameters. Unfortunately, it is very difficult to experimentally measure discrete bubbles parameters at high heat flux due to this bubble interaction process. For instance, in the HSV images, large vapor patches created by the coalesce of bubbles obstructs the field of view and do not allow to observe the near-wall boiling behavior. Only a few bubbles can be tracked. However, counting only individual (i.e., non-interacting) bubbles from HSV images may result in biases bubble size distribution (e.g., with smaller bubble size) at higher heat flux [98-99].

To understand the complex nature of boiling, thermal interaction between nucleation sites has been investigated, as illustrated in Figure 4.4. (a). Kenning and DelValle [100] analytically modeled bubble influence area and the overlap with inhibited nucleation zone. However, they concluded that a better understanding of bubble coalescence was required. Kitron et al. [101] analytically and stochastically modeled interaction between nucleation sites. Sadasivan et al. [102] developed a computational model to simulate boiling and identified the effects of activation and deactivation of nucleation sites on heat transfer. Zou and Jones [103] analyzed thermal interaction between nucleation sites by numerical simulation. Recently, Su et al. [54] stochastically modeled a fraction of active nucleation sites that can simultaneously sustain bubbles.

Hydrodynamic interaction between bubbles (see Figure 4.4. (b)) has been investigated as well. Chai [104] developed a theoretical framework to analyze bubble interaction. He presented mechanistic description to analytically capture the self-organizing nature of boiling. Zhang and Shoji [105] observed bubble coalescence on two artificial cavities with different spacing. They examined the effects of the bubble interaction on the bubble departure frequency. Likewise, investigation of bubble coalescence phenomena with a small number of artificial cavities has been conducted in many other studies [106-108]. Recently, Kim and Kim [109] mechanistically modeled coalesced bubble area by assuming a lattice of nucleation sites for HFP modeling. Their results showed constant two-phase area fraction at high heat flux.
Figure 4.4 Schematic diagram of various modeling strategies for dynamic interactions during boiling. (a) Modeling of the portion of active nucleation sites (b) Modeling of coalesced bubble area (c) Modeling of individual inherent nucleate bubble size (this study).

Although the importance of mechanistic modeling of bubble size distribution has been highlighted in other two-phase flow studies [110-112], little has been reported for boiling bubbles. In the boiling community, Klausner et al. [113] were concerned by predicting the probability density function of bubble size in pool and flow boiling conditions. They analytically modeled bubble departure and lift-off diameter distributions in flow boiling. Maurus et al. [72] pointed out that the physical importance of presenting bubble parameters as distribution functions rather than averaged value is to preserve information and avoid deterministic view in the boiling process.

In this study, an analytical model was developed to process experimental data and reconstruct the size distribution of potentially non-interacting, discrete bubbles, starting from the measured size distribution of discrete, but eventually coalescing bubbles, as illustrated in Figure 4.4 (c). The analytical model requires parameters such as distance between nucleation sites, bubble size distribution after coalescence, and bubble nucleation probability, which involve the information about thermal and hydrodynamic interactions between nucleation sites and bubbles (see Section 4.2.1). The non-linear analytical equations were numerically solved by implementing the stochastic gradient descent (SGD) method. We induced a new weight updating function for the proposed non-linear system. The mechanistic model was validated by stochastic modeling of synthetic boiling data (see Section 4.2.2). Finally, the model was applied to experimental data in
Section 4.2.3 to demonstrate its application at high heat flux.
Mechanistic modeling of individual bubble size distribution in this study will extend the experimental observation to practical applications, such as HFP modeling, CFD simulation [114] and validation of the existing bubble growth models [115].

4.2.1 Development of Analytical Model and Numerical Solver

When we assume a nucleation site \( i \) has its own inherent bubble radius distribution, \( f_{ri} \) (see Figure 4.5 (b)), the non-interaction probability of a bubble with radius \( k \) can be expressed as Eq. (4.12).

\[
f_{ri}(k) \cdot \Pi_{j=1,j \neq i}^{N} \left( 1 - P_{nuc,j} + P_{nuc,j} \cdot \int_{0}^{r_{ij}-k} f_{rj}(m)dm \right) \\
= f_{ri}(k) \cdot \Pi_{j=1,j \neq i}^{N} \left( 1 - P_{nuc,j} + P_{nuc,j} \cdot F_{rj}(l_{ij} - k) \right)
\]  

(4.12)

where \( f_{ri}(k) \) is the probability of a bubble having radius \( k \), \( N \) is the total number of nucleation sites, \( P_{nuc,j} \) is the probability of a nucleation site \( j \) to be activated, \( l_{ij} \) is the distance between nucleation sites \( i \) and \( j \) (see Figure 4.5 (a)), \( f_{rj}(m) \) is the probability of bubble nucleating at a site \( j \) and with a radius \( m \), and \( F_{rj}(l_{ij} - k) \) is the cumulative probability of a bubble nucleating at a site \( j \) and having radius below \( l_{ij} - k \). During boiling, the apparent individual (i.e., non-coalesced) bubble radius distribution of a site \( i \), \( f_{ri}^{boil} \), is different from \( f_{ri} \) due to interaction with neighboring bubbles (see Figure 4.5 (c)). The probability of an individual bubble having radius \( k \) is

\[
f_{ri}^{boil}(k) = \frac{f_{ri}(k) \cdot \Pi_{j=1,j \neq i}^{N} \left( 1 - P_{nuc,j} + P_{nuc,j} \cdot F_{rj}(l_{ij} - k) \right)}{\int_{0}^{r_{i,\text{max}}} f_{ri}(r_{i}) \cdot \Pi_{j=1,j \neq i}^{N} \left( 1 - P_{nuc,j} + P_{nuc,j} \cdot F_{rj}(l_{ij} - r_{i}) \right)dr_{i}}
\]  

(4.13)

where \( f_{ri}^{boil}, P_{nuc,j}, N, l_{ij}, \) and \( r_{i,\text{max}} \) can be obtained from experimental data (\( P_{nuc,j} \approx f_{i} \cdot t_{g,i} \), where \( f_{i} \) is nucleation frequency and \( t_{g,i} \) is growth time for site \( i \), and \( f_{ri} \) is the unknown. In order to facilitate processing experimental data and calculations, the denominator of Eq. (4.13) is discretized as:

\[
f_{ri}^{boil}(k) = \frac{f_{ri}(k) \cdot c_{lk}}{\sum_{k=0}^{r_{i,\text{max}}} f_{ri}(k) \cdot c_{lk}}
\]  

(4.14)
where $C_{i,k}$ is defined as,

$$C_{i,k} \equiv \prod_{j=1,j \neq i}^{N} \left( 1 - P_{nuc,j} + P_{nuc,j} \cdot F_{rj}(l_{ij} - k) \right)$$

Expanding Eq. (4.14) with the discretized radius gives

$$f_{r_i}^{\text{boil}}(k)f_{r_i}(0)C_{i,0} + f_{r_i}^{\text{boil}}(k)f_{r_i}(1)C_{i,1} + \cdots + (f_{r_i}^{\text{boil}}(k) - 1)f_{r_i}(k)C_{i,k} + \cdots + f_{r_i}^{\text{boil}}(k)f_{r_i}(r_{i,\text{max}})C_{i,r_{i,\text{max}}} = 0$$

(4.16)

**Figure 4.5** Conceptual diagram for a nucleation site $i$. (a) Effective bubble area of nucleation site $i$ and neighboring sites (b) Inherent nucleate bubble radius distribution (c) Apparent individual bubble radius distribution on boiling.

Note that, each nucleation site $i$ involves a set of equations similar to Eq. (4.16) for each discretized bubble radius value ranging from 0 to $r_{i,\text{max}}$. The sets of equations for the total number of nucleation sites, $N$, can be organized in a nonlinear sparse matrix, $ABx = 0$, as shown in Figure 4.6. The sparse matrix $A$ consists of known values, i.e., apparent individual bubble size distribution. The matrix $B$ is a block-diagonal matrix including unknowns, i.e., cumulative density function of nucleate bubble radius. The vector $x$ is the solution we want to know, i.e., the inherent probability density function of nucleate bubble radius. Since the matrix $B$ consists of the convoluted form of unknowns, the nonlinear system cannot be solved with existing nonlinear solvers. Therefore, a new methodology was proposed in this work.
We adapted the classical stochastic gradient descent (SGD) method. The schematic flow diagram of the algorithm is shown in Figure 4.7. The algorithm starts with the initial set of parameters, $x_0$, which represents the raw experimental data, i.e., the individual bubble size distribution. A linear module is used to update $x_1$ with a square weight matrix $W$.

$$x_1 = Wx_0$$

(4.17)

Then, $x_1$ is plugged in the nonlinear system to produce $x_2$,

$$x_2 = ABx_1$$

(4.18)

where $B$ is also a function of $x_1$. A Euclidean loss is calculated to update weights to minimize the error,

$$C(x_2) = \|x_2 - y\|^2$$

(4.19)

where $y$ is the desired value ($y = 0$ for our nonlinear system). The weight $W$ is updated by backpropagation,

$$W^{t+1} = W^t + \eta \frac{\partial C}{\partial W}$$

(4.20)

where $t$ is an iteration index and $\eta$ is a constant to control convergence rate. In this study, $\eta$ is set to be 1. Note that, the value of $\eta$ is several orders larger than the typical learning rate used when the SGD method is applied for training in machine learning problems ($<<1$). We need to compute gradients of the error with respect to the weight, which is essentially obtained through a backpropagation chain rule of derivatives. The transpose of the gradients of the error is given by

$$\left(\frac{\partial C}{\partial W}\right)^T = \frac{\partial x_2}{\partial W} \cdot \frac{\partial C}{\partial x_2}$$

$$= \frac{\partial x_2}{\partial x_1} \cdot \frac{\partial x_1}{\partial W} \cdot \frac{\partial C}{\partial x_2}$$

$$= AB \cdot x_0 \cdot x_2^T$$

$$= AB \cdot x_0 \cdot (ABWx_0)^T$$

(4.21)

The derivative of matrix can be calculated by Jacobian formulation. After iterations, when the error becomes smaller than a prescribed value (e.g., 1E-4), we obtain the inherent bubble size distribution for all nucleation sites $x_1$.

The performance of the proposed mechanistic model was validated by stochastic simulations of the boiling process, discussed in the following section (i.e., Section 4.2.2).
Figure 4.6 Nonlinear system of bubble size distribution for all nucleation sites.

\[ x_0 \rightarrow W \rightarrow x_1 \rightarrow F(x_1) \rightarrow x_2 \rightarrow C(x_2) \]

Initial input  Weight function  Linear module  Nonlinear system  Output  Cost function

\[ W^{t+1} \leftarrow W^t + \eta \frac{\partial C}{\partial W} \]

Figure 4.7 Schematic flow diagram of the proposed nonlinear solver.
4.2.2 Validation of the Mechanistic Model by Stochastic Simulations

Monte-Carlo (MC) technique has been used for stochastic modeling of boiling in previous studies [54, 101, 117-118]. In the current work, a stochastic simulation was used, to generate bubbles according to a given radius distribution and nucleation probability. Figure 4.8 (a) shows the maximum bubble occupancy for four nucleation sites. In order to validate the mechanistic model at high heat flux conditions, a high coalescence test case was considered. An exponentially dampened probability density function (PDF) was used for the radius at each nucleation site [117]. Figure 4.8 (b) shows initial input variables and MC simulation results after 10000 iterations. A gray line is an input size PDF according to which bubbles are generated on each of the four nucleation sites. At every time step, a nucleation site is considered to be active if a random number ([0 1]) is below a nucleation probability. For active nucleation sites, a bubble independently “nucleates” on each site following the input size PDF. If a bubble is an individual patch, i.e., does not interact with other bubbles, the radius was recorded. The output individual bubble PDF is denoted as ‘MC simulation’ in Figure 4.8 (b). Since larger bubbles coalesce easily and are excluded from the counting, the radius distributions of non-interacted individual bubbles shifts towards smaller values compared to the initial input.

The output of the MC simulation, i.e., individual bubble size distribution after interaction, was then used as an input to the mechanistic model for its validation. The objective of the model is to reconstruct inherent nucleate bubble size distribution for each site, which is equivalent to the ‘Initial input’ in Figure 4.8 (b). By knowing the solutions a priori, i.e., gray lines in Figure 4.8 (b), the accuracy of the model is qualitatively demonstrated. The radius distributions reconstructed by the model, ‘Reconstructed’ in Figure 4.8 (b), show good agreement with the initial nucleation bubble size distributions, ‘Initial input’, up to the measurable radius range after 2079 iterations. Figure 4.9 shows the averaged radius distribution over all nucleation sites. After interactions, the averaged individual bubble radius (12.7 pixel) is smaller than the initially given value (16.5 pixel). However, the reconstruction provides an average value (15.3 pixel) close the input value.

In conventional HFP modeling, the averaged radius over all nucleation sites is used. The discussion in this section implies that we may experience measurement biases when we use as-recorded experimental radius data. Processing experimental data and reconstructing inherent bubble nucleating characteristics for each nucleation site will improve the modeling accuracy.
Figure 4.8 Validation of the mechanistic model with MC simulation (a) MC simulation synthetic bubble configuration (b) MC simulation and reconstructed results.

Figure 4.9 Averaged distribution over four sites.
4.2.3 Application to Subcooled Flow Boiling at High Heat Flux

The mechanistic model was applied to phase-detection HSV experimental images in subcooled flow boiling (at 1 bar, 10 K subcooling, 1000 kg/m²s mass flux, on plain ITO) [55] to investigate individual bubble size distribution and to evaluate its applicability at high heat flux. The phase detection technique clearly discriminates different phases of a bubble, i.e., vapor, liquid microlayer, and dry area, as shown in Figure 4.10 (a). In particular, the existence of dry area indicates that bubbles are attached to the surface and contribute to near-wall heat transfer. The three phases were segmented using U-net [85] as shown in Figure 4.10 (b). A separate training set was created for each region. Equivalent radii of individual vapor bubbles and individual microlayers were measured, respectively. A bubble with multiple microlayer areas or dry areas was considered as a coalesced patch. Similarly, a microlayer with multiple dry area was considered as a coalesced patch. A bubble without dry area was excluded, as it had detached from the surface. The selected individual patches for the vapor bubble and microlayer are shown in Figure 4.10 (c).

Figure 4.10 Post-processing of phase-detection HSV (a) Raw image at 1610 kW/m² (b) Vapor bubble, microlayer bubble footprint, and dry area bubble base detection by U-net (c) Selected individual patches.
Since visualization of microlayer is a way to directly observe near-wall bubble formation, a microlayer provides better resolution to discriminate individual patches, e.g., a coalesced vapor bubble may contain separate individual microlayer and dry areas. The growth time, $t_g$, on each nucleation site was calculated as the time that a dry area lasts on the heated surface. The nucleation probability of each site was calculated as $P_{nuc} = f \cdot t_g$. Since the mechanistic model was not originally developed taking the interaction between sliding bubbles into account, a sliding length of an upstream bubble was added to its radius to simulate interaction with downstream bubbles.

The experimentally measured individual radius distribution was first fitted with a Gamma distribution. Original experimental data and the best Gamma fit for individual microlayer at 970 kW/m$^2$ are shown in Figure 4.11 (a). The averaged individual radius is plotted in Figure 4.11 (b) as a function of heat flux. The radius of individual microlayer is getting larger and becomes almost constant at high heat flux, which is consistent with the previous findings [54]. However, vapor bubbles are significantly coalesced at high heat flux, and only small size of individual bubbles survive without interacting. Therefore, the averaged radius for individual bubbles at high heat flux, e.g., at 2030 kW/m$^2$, was underestimated. This measurement bias of individual bubble size may lead to inaccurate HFP estimation at high heat flux.

The dotted lines in Figure 4.11 (a) and (b) indicate the reconstructed individual radius distribution and averaged value, respectively. In particular, reconstruction of the individual vapor bubble radius distribution at high heat flux reduces the measurement bias. The reconstructed average radius of vapor bubble shows trends similar to the microlayer, i.e., larger at higher heat flux (see Figure 4.11 (b)). The reconstructed radius indicates the inherent bubble size, to which a bubble will grow at the given heat flux if it does not interact with other bubbles.

In light of the current understanding, the mechanistic model developed in this study would eliminate the measurement bias arising from the limited experimental observation using shadowgraph technique. The proposed model can be incorporated in the post-processing stage to calculate the exact individual bubble quantities for next-generation high heat flux HFP modeling.
Figure 4.11 Reconstruction of the non-interacted state (a) Radius distribution (b) Averaged radius for all nucleation sites.
Chapter 5

Experimental Results and Analysis

5.1 Separate Effect of Surface Micro-Morphology on Subcooled Flow Boiling

In Section 1.1, we showed results of widely different subcooled flow boiling HTC and CHF on randomly polished samples with the same surface properties (e.g., average roughness, wettability) under the exact same operating conditions. Since the scratch pattern is the first noticeable difference between randomly polished samples (see Figure 1.1), we investigated the separate effect of groove direction on subcooled flow boiling to ascertain if the randomly created scratch pattern is the cause of the observed discrepancies.

A 1.015 ± 0.015 mm thick Zircaloy-4 heater was prepared by uni-directional hand polishing obtaining an average surface roughness $S_a$ of 715 ± 312 nm and contact angle of 54.8 ± 7.8°. A subcooled flow boiling experiment at 1 bar, 10 K subcooling, and with a 1000 kg/m²s mass flux was conducted with the groove direction spanwise, i.e., perpendicular to the flow. After the first test, the heater was rotated by 90° and the second experiment was conducted with the groove direction streamwise, i.e., parallel to the flow. Note that, the first boiling test including CHF did not change the surface microscopic features (detail in Appendix A). Since the horizontal and the vertical grooves are the two geometrical extremes regarding to the flow direction, any effect of the micro-morphology on subcooled flow boiling should be maximized. To prepare the sample for the second test (i.e., with vertical grooves), the heater layers were re-coated following the procedure described in Section 2.2 to preserve the boiling surface intact. The eventual heater thickness was 0.88 ± 0.01 mm.

Boiling curves and HSV images are shown in Figure 5.1. The shadowed area in a boiling curve represents the standard deviation of the spatial temperature distribution, as discussed in Section 3.1.3. It is clear that HTC, CHF, and bubble dynamics are the same for the two groove directions.
HSV images were post-processed using the algorithms described in Section 3.2 to measure fundamental boiling parameters (see Figure 5.2). Nucleation site density \((N'')\), bubble departure diameter \((D_d)\), growth time \((t_g)\), and nucleation frequency \((f)\) were practically the same for both the spanwise and streamwise grooves. These results suggest that the groove direction only may not affect the bubble dynamics or the entire boiling process as much as other parameters. Instead, the different boiling behavior observed on different fresh samples discussed in Section 1.1 could be due to the distribution of micron and sub-micron scale imperfections serving as bubble nucleation sites.

The same tests were conducted on another sample to prove reproducibility (see results in Appendix B.1).
**Figure 5.2** Measured boiling parameters for spanwise (red dots) and streamwise (black dots) grooves.
5.2 Effect of Substrate Thickness of Zircaloy-4 on Subcooled Flow Boiling

In order to avoid thickness driven bias on CHF, asymptotic CHF and asymptotic thickness of Zircaloy-4 in subcooled flow boiling were investigated. Figure 5.3 shows CHF of Zircaloy-4 at 1 bar, 10 K subcooling, and five mass flux conditions: 1000 kg/m²s, 750 kg/m²s, 500 kg/m²s, 360 kg/m²s, and 55 kg/m²s. The asymptotic CHF for each mass flux was determined as the best fit for the experimental data satisfying a $\propto G^{0.4}$ trend [119]. For 1000 kg/m²s, the measured average CHF was 2913 kW/m².

The pool boiling asymptotic thickness for SS and Inconel, $\delta_{asy, pool} = 0.55$ mm, is marked as a vertical line in Figure 5.3. SS and Inconel have the similar effusivity with Zircaloy-4 (see Figure 1.2). We did not observe any thickness effect at lower mass flux for a heater thicker than 0.55 mm, although further experiments at lower thickness may reveal the thickness related biases in these conditions.

![Figure 5.3 CHF on Zircaloy-4 at different mass fluxes in 1 bar, 10 K subcooling.](image)

Since there are no existing data for the asymptotic thickness of Zircaloy-4 in subcooled flow boiling, pool boiling normalized CHF of SS and Inconel are compared in Figure 5.4. Experimental conditions of each dataset are tabulated in Table 5.1. As shown in Figure 5.4, no thickness driven bias on CHF was observed for a Zircaloy-4 heater thicker than 0.55mm at 1 bar, 10 K subcooling,
and 1000 kg/m²'s mass flux. Therefore, when we fabricated a Zircaloy-4 heater, especially during multiple polishing steps, we paid attention to keep the sample thicker than 0.55 mm. Note that the typical fuel rod cladding thickness for PWR is 0.572 mm [81].

Figure 5.4 Normalized CHF for Zircaloy-4, SS, and Inconel.

Table 5.1 Experimental conditions for the data given in Figure 5.4.

<table>
<thead>
<tr>
<th></th>
<th>Material</th>
<th>Working fluid</th>
<th>Boiling</th>
<th>Asymptotic CHF</th>
</tr>
</thead>
<tbody>
<tr>
<td>This study</td>
<td>Zircaloy-4</td>
<td>Water</td>
<td>Flow</td>
<td>2913 kW/m²</td>
</tr>
<tr>
<td>Tachibana (1967) [13]</td>
<td>SS</td>
<td>Water</td>
<td>Pool</td>
<td>1170 kW/m²</td>
</tr>
<tr>
<td>Golobic (1997) [17]</td>
<td>SS</td>
<td>FC-72</td>
<td>Pool</td>
<td>135 kW/m²</td>
</tr>
<tr>
<td>Carne (1965) [12]</td>
<td>Inconel</td>
<td>n-propanol</td>
<td>Pool</td>
<td>450 kW/m²</td>
</tr>
</tbody>
</table>
5.3 Separate Effect of Zircaloy-4 Oxidation on Subcooled Flow Boiling

5.3.1 Oxidation kinetics

Zirconium oxide has distinct characteristic features depending on the oxide weight gain, also known as pre-transition, transition, and post-transition stages. Oxide weight gain was measured on a daily basis to check the oxidation stage, as shown in Figure 5.5 (left). Weight gain was converted into oxide layer thickness according to a $1\ \mu\text{m} \text{ per } 14.9\ \text{mg/dm}^2$ proportionality [23]. A total of three samples underwent oxidation. Sample 0 was oxidized up to 19 days to confirm the post-transition stage. Sample 1 and Sample 2 were oxidized up to 15 days (point D) and taken out at point A, B, C, and D for surface characterization and subcooled flow boiling experiments. Error bars in Figure 5.5 (left) represent the standard deviation of the oxide thickness among the three samples. In the following sections, studies on Sample 1 are discussed. Separate tests conducted on Sample 2 are presented in Appendix B.2 to demonstrate the reproducibility of the results.

Point A indicates a fresh, unoxidized surface. Point B is in the pre-transition stage, where the thickness gain exhibits a less-than-linear growth consistent with the Baker-Just correlation [24] (see eq. (1.9)). Point C is in the transition stage, where the thickness gain follows a cyclic growth rate. The surface of the sample at point C is covered by a mixture of black and white oxide. Point D is in the post-transition stage, as evidenced by a linear growth rate. The sample at point D appears to be entirely covered by a white oxide. The oxide weight gain measurement at point B, C, and D were verified using a FEI Helios NanoLab™ 600 Dual Beam Focused Ion Beam (FIB) and showed good agreement.
Figure 5.5 Oxide thickness gain (left) obtained through weight gain measurements and corresponding surface appearance and thickness measurement with a FIB at each stage (right).
5.3.2 Surface characterization

Surface characterization at the fresh, pre-transition, transition, and post-transition stages was done following the procedures described in Section 2.4. Micro- and nano-scale topography and morphology on the exact same area, and wettability and wickability of Sample 1 are shown in Figure 5.6.

![Surface characterization on Sample 1.

Figure 5.6 Surface characterization on Sample 1.](image-url)
For the pre-transition oxide, oxidation improves the wettability while preserving the micro- and nano-scale surface features. For the transition oxide, different phase of oxide growth results in non-uniform white oxide formation and increased local micro-scale roughness. For the post-transition oxide, the micro-scale surface profile changes considerably due to the volume expansion resulting from the non-uniform oxide growth. Accumulation of local compressive stresses creates cracks initiated from the highly oxidized spots. On the other hand, oxidation affects the nano-scale surface profile in a different way. We can observe from AFM data that oxidation smoothens out the nano-scale structures. It is evident in the SEM images that a certain cavity is closing as the surface becomes oxidized. In summary, the existing cavities are slowly closing until the transition stage, but when the thick oxide layer begins to crack, in the post-transition stage, large cavities form. In particular, due to the micro-cracks (see Figure 5.7 (b)), there is wicking flow on the post-transition oxide, as shown in Figure 5.7 (a). Note that this surface has an apparent non-zero contact angle (see Figure 5.6) because it is actually hemi-wicking.

Figure 5.7 Hemi-wicking on the post-transition oxide (a) Wicking front and contact line (b) Zoomed-in micro-crack.

Figure 5.8 (a) shows micro-topography and contact angle for the fresh sample A (cf. Figure 1.1), whose boiling behavior is quite different from fresh Sample 1 (see Figure 5.9). Note that the difference in the surface profile between the two fresh samples is much smaller than the micro-scale alternation due to transition and post-transition oxides on an identical sample (see Figure 5.8 (b)).
**Figure 5.8** Surface characteristics on different samples (a) Sample A in Figure 1.1 (b) Surface profile distribution for the Sample 1 and Sample A.
5.3.3 Subcooled flow boiling

The boiling curves of fresh and pre-transition, transition, and post-transition oxides of Sample 1, and fresh Sample A are shown in Figure 5.9. The shadowed area in a boiling curve represents the standard deviation of the spatial temperature distribution, as discussed in Section 3.1.3. The heater thicknesses, which were carefully controlled to be at least 0.55 mm, and CHF values are tabulated in Table 5.2.

![Boiling curves of fresh and oxidized Zircaloy-4](image)

**Figure 5.9** Subcooled flow boiling experiments on fresh and oxidized Zircaloy-4 for different oxidation level.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Thickness (mm, ±0.01)</th>
<th>CHF (kW/m², ±10)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample A</td>
<td>Fresh: 0.72</td>
<td>2564</td>
</tr>
<tr>
<td></td>
<td>Fresh: 0.98</td>
<td>3112</td>
</tr>
<tr>
<td></td>
<td>Pre-transition: 0.88</td>
<td>3203</td>
</tr>
<tr>
<td></td>
<td>Transition: 0.75</td>
<td>3218</td>
</tr>
<tr>
<td></td>
<td>Post-transition: 0.65</td>
<td>3619</td>
</tr>
<tr>
<td>Sample 1</td>
<td>Fresh: 0.98</td>
<td>3112</td>
</tr>
<tr>
<td></td>
<td>Pre-transition: 0.88</td>
<td>3203</td>
</tr>
<tr>
<td></td>
<td>Transition: 0.75</td>
<td>3218</td>
</tr>
<tr>
<td></td>
<td>Post-transition: 0.65</td>
<td>3619</td>
</tr>
</tbody>
</table>

**Table 5.2** Heater thickness and CHF value for the data given in Figure 5.9.
HSV bubble dynamics were recorded for 2000 frames with 10000 fps at each heat flux level from onset of nucleate boiling (ONB) to 1800~2000 kW/m² with 100 kW/m² intervals. HSV snapshots at four selected heat fluxes are shown in Figure 5.10.

![Figure 5.10 HSV images for the data given in Figure 5.9.](image)

Two fresh samples (1 and A) have large differences in HTC, CHF, and bubble dynamics. These discrepancies are much bigger than the changes due to oxidation on the identical sample. These results demonstrate that a careful control of boiling surface conditions is essential to conduct separate effect study of surface oxidation in subcooled flow boiling heat transfer.

For the pre-transition oxide, ONB, HTC, and CHF are practically the same as the fresh surface. These observations demonstrated the reduced impact of wettability in subcooled flow boiling, compared to pool boiling. In previous pool boiling studies, up to 40 % CHF enhancement was reported on the pre-transition Zircaloy-4 oxide, and this effect was attributed to the increase of
wettability.

For transition oxide, in spite of the increased local micro-scale roughness, no noticeable change in boiling behavior was observed. The waviness, i.e., S-shape, of the boiling curve is presumably be due to the fluctuation of inlet subcooling (± 1 K), as discussed in Section 2.3.

For the post-transition oxide, HTC is still consistent, but CHF is enhanced by 16 %. Micro-cracks and wicking flow may play a crucial role. In order to quantitatively analyze bubble dynamics, we post-processed HSV data with the originally developed algorithms described in Section 3.2.

Individual bubble parameters (e.g., departure diameter, growth time, nucleation frequency, lift-off maximum axis diameter, and sliding length) were measured for each nucleation site. Figure 5.11 (a)-(c) show bivariate PDFs of the measured quantities for all the nucleation sites from ONB to the maximum heat flux where HSV were recorded. The bubble departure diameter shows an inverse relation with the nucleation frequency, whereas it shows a proportional relation with growth time (see Figure 5.11 (a)-(b)). Bubble lift-off diameter and sliding length have a proportional relation as well (see Figure 5.11 (c)). Dotted lines in Figure 5.11 (a)-(c) represent the averaged values for all the heat fluxes. Figure 5.11 (d) shows the nucleation site density as a function of heat flux.

Fundamental boiling parameters for the fresh, pre-transition oxide, and transition oxide are distributed analogously. However, for the post-transition oxide, the boiling parameter distributions tend to cluster towards smaller bubble size and short growth/sliding time, as shown in Figure 5.11 (b) and (c). These trends can be explained by the surface modification due to long-term oxidation as discussed in Section 5.3.2. As sub-micron structures are smoothened and existing cavities are closed, newly opened cavities due to micro-cracks dominate the active nucleation site distribution. The new active cavity distribution may have a slightly larger size than the average size of the previously existing cavities. Therefore, bubble size distribution on the post-transition oxide becomes less dispersed and clustered within a smaller size range. Since micro-cracks spread over the boiling surface, more cavities will be activated at the same wall superheat (i.e. higher nucleation site density) compared to the “before-post-transition” stages, as shown in Figure 5.11 (d), because larger size cavities are easier to activate. Meanwhile, we can speculate that hemi-wicking may promote a bubble to depart earlier from the surface, so that an individual bubble travels less distance during sliding (see Figure 5.11 (c)). The longer wait time offset by the shorter growth time makes the nucleation frequency smaller, as shown in Figure 5.11 (a).
Figure 5.11 Quantified fundamental boiling parameters.
The measured bubble parameters were input into the heat flux partitioning model discussed in Section 4.1, as shown in Figure 5.12. Two different modeling strategies are compared. Figure 5.12 (a) is the approach proposed in this study, i.e., using quantities at each nucleation site for site-wise summation and considering asymmetric microlayer growth. Figure 5.12 (b) is following the conventional strategy, i.e., using averaged parameters for all nucleation sites and assuming symmetric growth. In Figure 5.12 (b), the fraction of active nucleation sites was calculated by CSR approximation [92]. The value of averaged parameters is listed in Appendix C. The proposed HFP modeling in Figure 5.12 (a) does not require additional averaging steps which may create another uncertainty in handling parameters. Due to shorter bubble sliding distance on post-transition oxide, sliding conduction and sliding evaporation take a small portion (see Figure 5.12 (a), post-transition). We can speculate that the portion of static quenching heat flux will be increased at high heat flux on post-transition oxide, as extrapolated from Figure 5.12 (a). Although static evaporation is the powerful heat removal mechanism, it promotes merging of bubbles at high heat flux. Contrastively, the enhanced static quenching due to wicking flow can be the potential mechanism leading to the higher CHF on post-transition oxide.

**Figure 5.12** Heat flux partitioning by two different modeling strategies. (a) The model proposed in this study; Use quantities at each nucleation site for site-wise summation and consider asymmetric microlayer growth (b) The conventional method; Use averaged parameters for all nucleation sites and assume only symmetric growth.
5.4 Separate Effect of ATF Cladding Material on Subcooled Flow Boiling

5.4.1 Surface characterization

Surface characterization for Zircaloy-4 and FeCrAl heaters before and after Cr-coating was done following the procedures described in Section 2.4. Micro- and nano-scale topography and morphology on the exact same area before and after Cr-coating and wettability are shown in Figure 5.13.

Although Zircaloy-4 and FeCrAl samples were simultaneously and uni-directionally polished, their surface finish was different. The FeCrAl surface is rougher and has more narrow peaks and valleys than Zircaloy-4, as revealed by the average micro-roughness and the nano-topography (AFM data). The higher roughness ratio on FeCrAl (r=2.11) is due to the more rugged nano-scale structure compared to Zircaloy-4 (r=1.03). These differences are presumably due to distinct mechanical properties (e.g., hardness) of Zircaloy-4 and FeCrAl. Due to the larger peak structures on FeCrAl, the contact line pins and the contact angle is higher (note that the contact angle was measured from images perpendicular to the grooves direction (i.e. parallel to the flow)).

After running subcooled flow boiling experiments on fresh Zircaloy-4 and FeCrAl, each surface was coated with Cr by PVD. Cr-coating conformally covered the surface, smoothening the nano-scale features and preserving the micro-scale structures. A noticeable sub-micron transformation on FeCrAl can be observed in the AFM data where, after the Cr coating, the roughness ratio decreased significantly and became similar to that of the Cr-coated Zircaloy-4. SEM imaging on both Cr-coated Zircaloy-4 and FeCrAl clearly demonstrates the effects of PVD Cr-coating. The Cr-coating modifies the FeCrAl nano-scale morphology as if the surface is covered with “snow” that makes the nano-morphology of Zircaloy-4 and FeCrAl similar. The overall observation aligns well with the previous study of Umretiya et al. [36]. They argued that the sub-micron rounded shape features enhance wetting, so that the PVD Cr-coated surface has lower contact angle (i.e., more hydrophilic).
Figure 5.13 Surface characterization on Zircaloy-4 and FeCrAl before and after Cr-coating.
5.4.2 Subcooled flow boiling

The boiling curves of fresh and Cr-coated Zircaloy-4 and FeCrAl are shown in Figure 5.14. The shadowed area in a boiling curve represents the standard deviation of the spatial temperature distribution, as discussed in Section 3.1.3. The wavy shape of the boiling curve is presumed to be due to the fluctuation of inlet subcooling (±1 K) (discussed in Section 2.3). Heater thicknesses, which safely exceed asymptotic thickness limit, and CHF values are tabulated in Table 5.3.

![Figure 5.14 Subcooled flow boiling experiments on fresh and Cr-coated Zircaloy-4 and FeCrAl](image)

<table>
<thead>
<tr>
<th></th>
<th>Fresh Zircaloy-4</th>
<th>Cr-coated Zircaloy-4</th>
<th>Fresh FeCrAl</th>
<th>Cr-coated FeCrAl</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thickness (mm, ±0.01)</td>
<td>0.98</td>
<td>0.8</td>
<td>0.92</td>
<td>0.79</td>
</tr>
<tr>
<td>CHF (kW/m², ±10)</td>
<td>3175</td>
<td>2858</td>
<td>3184</td>
<td>2806</td>
</tr>
</tbody>
</table>

Table 5.3 Heater thickness and CHF value for the data given in Figure 5.14.
HSV bubble dynamics were recorded for 2000 frames with 10000 fps at each heat flux level from onset of nucleate boiling (ONB) to 1500~1700 kW/m$^2$ at 100 kW/m$^2$ intervals. HSV snapshots at the selected four heat fluxes are shown in Figure 5.15.

**Figure 5.15** HSV snapshots for the data given in Figure 5.14.

The post-processed HSV data, i.e., quantified fundamental boiling parameters, are plotted in Figure 5.16 (a)-(d) in the same way they were presented for the oxidation study in Section 5.3.3. The partitioned heat fluxes are shown in Figure 5.17. For fresh FeCrAl (see Figure 5.17 (c)), HSV data were collected until ~1500 kW/m$^2$ similar to other surface.
Figure 5.16 Quantified fundamental boiling parameters.
Fresh FeCrAl has earlier ONB (see Figure 5.14), i.e., it starts to boil at lower wall superheat than fresh Zircaloy-4, presumably due to the large size or reentrant shape of active cavities, which are expected given the higher roughness ratio. Since larger cavities have a lower nucleation temperature, bubbles tend to grow slow (compared to bubble nucleating at higher temperature). These slowly growing bubbles tend to depart earlier from the surface (with little or no sliding), i.e., an individual bubble on FeCrAl travels a shorter distance and lifts off from the surface earlier than on fresh Zircaloy-4, as shown in Figure 5.16 (c). As bubbles are smaller (see Figure 5.16 (a) and (b)), more nucleation sites are active on the heated surface, i.e., FeCrAl has a higher nucleation site density compared to the other surfaces (see Figure 5.16 (d)). The results show good agreement with the pool boiling study on monolithic FeCrAl alloy [41], where FeCrAl has higher average surface roughness, enhanced HTC, and CHF similar to Zircaloy-4.

Cr-coated Zircaloy-4 and Cr-coated FeCrAl have a similar ONB temperature as fresh Zircaloy-4. The Cr-coating conformally covered the rough features of FeCrAl making its nanostructure similar to Zircaloy-4 and the Cr Coated Zircaloy-4. This also increase the ONB temperature and, accordingly, the entire bubble dynamics, e.g., causing larger bubbles (see Figure 5.16 (a) and (b)). Since larger bubble growth may de-activate neighboring nucleation sites, Cr-coated FeCrAl surfaces have smaller nucleation site density, as shown in Figure 5.16 (d). This combination results in a lower HTC and CHF. The results seem to contradict previous pool boiling study on DC sputtered Cr-coating on SS [40], showing that a Cr-coating makes the surface super-hydrophilic and enhance CHF. We speculate that the effect of nanostructures (that reduces CHF) is more influential than the effect of wettability (that enhances CHF) in subcooled flow boiling.

Results of a second series of tests for fresh Zircaloy-4 and fresh FeCrAl are shown in Appendix B.3.
Chapter 6

Conclusions

6.1 Summary of the observations

In this study, the separate effects of surface morphology, substrate thickness, Zircaloy-4 oxidation, and ATF material on subcooled flow boiling have been investigated. To that end, a specialized heater fabrication procedure (e.g., polishing, sputtering, oxidation) was developed to carefully control surface finish conditions. A unique surface characterization technique was established to measure micro- and nano-scale topography and morphology on the exact same surface region for an identical sample. Subcooled flow boiling experiments were conducted at 1 bar, 10 K subcooling, and 1000 kg/m²s mass flux conditions with high-resolution HSV and IR cameras. A STAR-CCM+ 3-D inverse conduction model as well as MATLAB 1-D inverse conduction algorithm were developed to obtain wall superheat and heat flux from measurement of the temperature and heat flux on the airside of the boiling sample. A machine learning tool and an optical flow algorithm were implemented to post-process the HSV images and measure fundamental boiling parameters. A modified heat flux partitioning model was proposed accounting for the non-symmetric growth of the micro-layer in presence of the flow.

The separate effect investigation on surface morphology revealed that scratch pattern-morphology (i.e., groove direction) does not affect subcooled flow boiling. Instead, it suggests that the formation of nucleation sites, e.g., their location, size, and shape, is the most dominant factor to determine the outcome of the boiling process.

The asymptotic CHF and thickness studies revealed that no thickness bias on CHF was observed for a Zircaloy-4 heater thicker than 0.55 mm on subcooled flow boiling.

The separate effect investigation on Zircaloy-4 oxidation showed that no changes in HTC and CHF on pre-transition and transition oxides, where wettability and micro-roughness were increased. On the other hand, CHF was enhanced on post-transition oxide where micro-cracks created large size of nucleation sites and wicking flow. These findings seem to disagree with the conclusions drawn in previous researches, where the enhancement of pool boiling CHF on pre-transition oxide was
attributed to the improvement of the surface wettability. Instead, in subcooled flow boiling, more important factors are the formation of active nucleation cavities and the overall surface morphology.

The separate effect investigation on ATF material revealed that rougher structures on fresh FeCrAl lead to lower ONB and higher HTC with smaller bubbles and lower nucleation site density compared to fresh Zircaloy-4. Nano-scale morphology modification by PVD Cr-coating leads to reduced HTC and CHF. These findings are different from the common understanding in previous pool boiling research, where higher wettability generally gives enhanced CHF. Although wettability and average micro-roughness are considered to be important properties in pool boiling, micro- and nano-scale surface features are the dominant factors to change bubble dynamics and determine CHF and HTC in subcooled flow boiling.

6.2 Original Contributions

6.2.1 Scientific contributions

- Wettability and averaged surface roughness are not good indicators of the subcooled flow boiling performance. Instead, one should examine the surface structure at both the micro- and nano-scale and how the surface preparation process affect the formation of nucleation cavities. The findings in this study seem to corroborate the argument made in some previous pool boiling studies that averaged surface roughness is not a good indicator, but the number of nucleation sites and their size are the important factors to determine pool boiling HTC [1-3, 7, 10].

- When investigating surface oxidation effects, the finish of the fresh, un-oxidized surface (i.e., the distribution of nucleation cavities) is the primary factor to determine the efficiency of the subcooled flow boiling process. Boiling on different fresh samples (with the same wettability and averaged micro-roughness) may lead to larger discrepancies in HTC, CHF, and bubble dynamics than an identical sample before and after surface aging (e.g., oxidation). In light of these observations, we may have to re-consider some previous literature results where multiple samples were used as a control group, so that effects of
oxidation were not properly separated from the effects of surface finish conditions.

6.2.2 Technical contributions

The techniques and models developed in this study will be used for future separate effect boiling heat transfer investigations as well as analysis of experimental data (e.g., HSV images). For example:

- An original metallic heater design resembling nuclear fuel cladding heating configuration and the systematic heater fabrication procedure were introduced.
- A unique methodology to complete systematic surface characterization was introduced: complementary multiple-scale (i.e., from nano- to micro-scale) measurements on an identical area before and after surface modification.
- 1-D/3-D inverse conduction solvers were developed using MATLAB and STAR-CCM+ to obtain the temperature and heat flux at the boiling surface with thick metallic heaters.
- HSV post-processing methodology including U-net and optical flow algorithms was developed to extract quantified boiling parameters from HSV data.
- A modified HFP model incorporating the modeling of non-uniform micro-layer growth was proposed.
- An analytical model and a numerical non-linear solver to reconstruct inherent nucleate bubble size distribution were proposed for next-generation high heat flux HFP application. Calculation of partitioned heat fluxes using shadowgraph technique on metallic heater can be extended to higher heat flux with the modeled individual bubble size.
6.3 Suggested Future Work

6.3.1 Artificial CRUD deposition

In PWRs, CRUD deposition naturally changes cladding surface properties [33, 120-123]. Separate effect investigation on CRUD deposition will provide significant findings to understand subcooled flow boiling behavior in nuclear reactor. To that end, an experimental facility for artificial CRUD deposition and a test scheme are suggested hereafter.

Figure 6.1 (a) shows a tube pool boiling set-up. The bottom of the tube is sealed with an o-ring pushed by the top SS support structure. The copper block heater at the bottom has a contact area of $1 \times 1$ cm$^2$ with a Zircaloy-4 sample. A $2 \times 2$ cm$^2$ Zircaloy-4 sample can be soldered on the copper heating block, and the active boiling area becomes $1 \times 1$ cm$^2$. We created the artificial CRUD solution similar to reactor coolant components, which consists of 50 ppm of Fe$_2$O$_3$, 50 ppm of NiO, 500 ppm of H$_3$BO$_3$, and 2 ppm LiOH. The appearance of a Zircaloy-4 sample deposited with 1~2 $\mu$m thick artificial CRUD is shown in Figure 6.1 (b). CRUD forms around active nucleation sites. Figure 6.2 shows SEM images of the CRUD layer, which clearly demonstrate the porous features.

![Figure 6.1 Artificial CRUD deposition on Zircaloy-4](a) Artificial CRUD pool boiling deposition facility (b) Artificial CRUD deposited Zircaloy-4.
Figure 6.2 Artificial CRUD SEM images obtained on the sample in Figure 6.1 (b).

Figure 6.3 shows the suggested test scheme for the future research to complete separate effect investigation on artificial CRUD deposition on Zircaloy-4 cladding on subcooled flow boiling.

Figure 6.3 Suggested test scheme for the separate effect investigation on artificial CRUD deposition on subcooled flow boiling.

6.3.2 Elevated pressure

In order to fully understand subcooled flow boiling heat transfer in PWRs, separate effect studies of Zircaloy-4 oxidation and ATF cladding material should be extended to high-pressure experiments, especially up to the reactor operating pressure (e.g., 150 bar). Although bubble dynamics are hard to be directly observed at elevated pressure due to technical limitations, e.g., very tiny bubbles are not visible in HSV cameras, methodologies and models developed in this study can be foundation works for the systematic separate effect investigation at high-pressure applications.
6.3.3 Nucleation site size distribution

One key take-away point in this study is that the size distribution of nucleation sites seems to be what matters the most to subcooled flow boiling performance. In order to demonstrate this hypothesis, techniques should be improved to characterize the size of real active cavities on randomly polished metallic surface. Enhanced spatial resolution of HSV camera will facilitate identifying accurate nucleation site locations. The identified nucleation sites further can be characterized using SEM for their size and shape. In this way, the multiple fresh samples having different boiling performance (see Figure 1.1 and 1.2) will exhibit distinct size distribution of active nucleation sites.

6.3.4 Re-visit pool boiling tests

Since our current claims as aforementioned in Section 6.2.1 have some disagreements with previous pool boiling studies, we can re-visit pool boiling tests, and confirm or dispute the findings in the current subcooled flow boiling tests.
Appendix A

Separate Effect of Subcooled Flow Boiling on Surface Condition

The test schemes proposed in this study (see Figures 1.7, 1.8, and 5.1) require multiple boiling tests on an identical sample for the separate effect investigation. Therefore, the effect of a boiling test (including CHF) on surface condition was examined beforehand. In several previous studies [7, 43], surface morphology was compared before and after boiling tests, and people concluded that surface is modified due to aging (e.g., erosion, deposition, oxidation, and corrosion) during boiling. However, their region of measurement was randomly chosen, which does not exclude the possibility that original differences exist on different regions of the fresh surface. In this study, surface characterization was done on the exact same surface region before and after boiling. Figure A.1 shows surface characterization results on two samples with different roughness. The micro-profile measurements display that there was little change in the microscopic properties during boiling. Meanwhile, surface became more hydrophilic due to aging. In subcooled flow boiling, the influence of wettability on HTC, CHF, and bubble dynamics diminishes compared to pool boiling. Therefore, we can conclude that a subcooled flow boiling test does not change surface condition.

![Surface characterization results](image)

**Figure A.1** Surface characterization before and after boiling.
Appendix B

Repeat Tests

In order to provide reproducibility, repeated tests were conducted on a second sample for each of the separate effect investigations of surface micro-morphology, Zircaloy-4 oxidation, and ATF cladding material. Boiling curves, HSV images, and surface characterization results are presented in the current section.

B.1 Surface Micro-Morphology

Figure B.1 Subcooled flow boiling experiments on Sample 2 for surface micro-morphology studies.
B.2 Zircaloy-4 Oxidation

Figure B.2 Surface characterization on Sample 2 for oxidation studies.
Table B.1 Heater thickness and CHF value for the data given in Figure B.3.

<table>
<thead>
<tr>
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<th>Sample 2</th>
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<tbody>
<tr>
<td></td>
<td>Fresh</td>
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<tr>
<td>Thickness (mm,  ±0.01)</td>
<td>0.97</td>
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<tr>
<td>CHF (kW/m²,  ±10)</td>
<td>2989</td>
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</table>
Figure B.4 HSV snapshots for the data given in Figure B.3.
B.3 ATF Cladding Material

**Figure B.5** Surface characterization on fresh Zircaloy-4 (Sample 2) and FeCrAl (Sample 2).

**Figure B.6** Subcooled flow boiling experiments on fresh and Cr-coated Zircaloy-4 and FeCrAl.
Table B.2 Heater thickness and CHF value for the data given in Figure B.6.

<table>
<thead>
<tr>
<th>Thickness (mm, ±0.01)</th>
<th>Fresh Zircaloy-4</th>
<th>Fresh FeCrAl</th>
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<tr>
<td>CHF (kW/m², ±10)</td>
<td>0.91</td>
<td>0.75</td>
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<td>3366</td>
<td>3263</td>
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</table>

Figure B.7 HSV snapshots for the data given in Figure B.6.
Appendix C

Averaged Boiling Parameters Used for The Conventional HFP Approach

Table C.1 Boiling parameters for the fresh surface.

<table>
<thead>
<tr>
<th>$\Delta T$ (°C)</th>
<th>$q''$ (MW/m$^2$)</th>
<th>$N^*$ (#)</th>
<th>$D_d$ (mm)</th>
<th>$f$ (1/s)</th>
<th>$t_g$ (ms)</th>
<th>$D_l$ (mm)</th>
<th>$l_s$ (mm)</th>
<th>$t_s$ (ms)</th>
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<td>3</td>
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<td>413.25</td>
<td>0.49</td>
<td>0.72</td>
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<td>0.56</td>
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<td>0.56</td>
<td>413.51</td>
<td>0.50</td>
<td>0.76</td>
<td>0.65</td>
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<td>0.86</td>
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<td>0.74</td>
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<td>0.76</td>
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<td>0.99</td>
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<td>0.72</td>
<td>457.32</td>
<td>0.51</td>
<td>0.73</td>
<td>0.69</td>
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<td>0.63</td>
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<td>0.65</td>
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<td>0.69</td>
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<td>26.98</td>
<td>1.64</td>
<td>102</td>
<td>0.56</td>
<td>460.49</td>
<td>0.42</td>
<td>0.59</td>
<td>0.72</td>
<td>0.25</td>
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<td>27.45</td>
<td>1.83</td>
<td>146</td>
<td>0.56</td>
<td>413.41</td>
<td>0.41</td>
<td>0.61</td>
<td>0.75</td>
<td>0.26</td>
</tr>
</tbody>
</table>

* $N_b'' = ft_gN''', N_a''' = e^{-N_b''\pi(D_d/2)^2}$. $N_a'''$ was used for the calculation.

Table C.2 Boiling parameters for the pre-transition oxide surface.

<table>
<thead>
<tr>
<th>$\Delta T$ (°C)</th>
<th>$q''$ (MW/m$^2$)</th>
<th>$N$ (#)</th>
<th>$D_d$ (mm)</th>
<th>$f$ (1/s)</th>
<th>$t_g$ (ms)</th>
<th>$D_l$ (mm)</th>
<th>$l_s$ (mm)</th>
<th>$t_s$ (ms)</th>
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<tr>
<td>18.26</td>
<td>0.48</td>
<td>10</td>
<td>0.50</td>
<td>257.69</td>
<td>0.47</td>
<td>0.55</td>
<td>0.44</td>
<td>0.22</td>
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<td>20</td>
<td>0.54</td>
<td>14</td>
<td>0.53</td>
<td>316.05</td>
<td>0.48</td>
<td>0.60</td>
<td>0.48</td>
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<td>22.02</td>
<td>0.64</td>
<td>19</td>
<td>0.57</td>
<td>362.29</td>
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<td>0.59</td>
<td>0.48</td>
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<td>23.8</td>
<td>0.75</td>
<td>26</td>
<td>0.58</td>
<td>369.28</td>
<td>0.48</td>
<td>0.60</td>
<td>0.51</td>
<td>0.23</td>
</tr>
</tbody>
</table>

120
Table C.3 Boiling parameters for the transition oxide surface.

<table>
<thead>
<tr>
<th>$\Delta T$ ($^\circ$C)</th>
<th>$q''$ (MW/m$^2$)</th>
<th>$N$ (#)</th>
<th>$D_d$ (mm)</th>
<th>$f$ (1/s)</th>
<th>$t_g$ (ms)</th>
<th>$D_l$ (mm)</th>
<th>$l_s$ (mm)</th>
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Table C.4 Boiling parameters for the post-transition oxide surface.

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<th>$q''$ (MW/m$^2$)</th>
<th>$N$ (#)</th>
<th>$D_d$ (mm)</th>
<th>$f$ (1/s)</th>
<th>$t_g$ (ms)</th>
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Bibliography


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