

Development of Surface Modification Techniques for Enhanced Safety of Light Water Reactors: Recent Progress and Future Direction at THLAB

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1. Introduction

Surface conditions have a significant impact on boiling heat transfer phenomena including boiling crisis, which is also called critical heat flux (CHF). Conventionally, it is reported that enhanced CHF was obtained on a wettable surface [1]. Moreover, since roughness is generally related to the presence of micro-cavities, improved boiling performance is expected for a surface with a higher roughness value [2; 3]. Recently, notable changes in boiling heat transfer have been reported through surface modification. For instance, during nanofluid boiling, development of a hydrophilic and porous layer on a heating surface was observed by Kim et al. [4]. They concluded that the CHF enhancement in nanofluid boiling was mainly affected by the surface characteristics of the developed layer.

Furthermore, an introduction of surface modification can be utilized to secure the safety of nuclear reactor systems. At many components of the reactor systems, energetic boiling heat transfer occurs, and potential thermal attack to the systems is expected under normal or accident environments. In particular, during a reactor operation, fission energy is deposited in the fuel assemblies in a core. Also, under severe conditions, failure of a reactor vessel may occur by high temperature molten materials. Thus, with a well-established surface favorable for boiling heat transfer, improved thermal margin in the nuclear system can be achievable.

Recently, the Nuclear Thermal-Hydraulic Laboratory (THALB) at Hanyang University has developed two distinct cutting edge techniques for the surface modification, which is potentially applicable to the nuclear system: sputtering and layer-by-layer (LbL) deposition techniques. In this article, we introduce the surface modification techniques and recent achievements. After a brief description of each deposition mechanism, an assessment of thermal margin for both the technologies is discussed based on pool boiling experiments conducted at THLAB. Moreover, in the latter part of each chapter, experimental facilities for applied heat transfer tests to consider reactor environments are presented.

2. Cutting Edge Techniques for Engineered Surfaces: Sputtering and Layer-by-Layer Deposition

The research direction of the modification techniques at THLAB has been categorized into three regions based on the system scale as shown in Fig. 1: two small scale regions and one larger scale region. At the small scale regions, modification for a fuel cladding and fuel assembly is mainly dealt with. On the other hand, the large scale region is concerned with large area surfaces of the safety system, such as an external reactor vessel, core catcher body, or spent fuel assemblies in the storage pool.

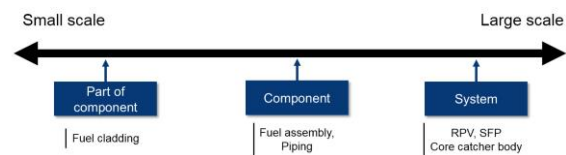


Fig. 1. Research ranges and corresponding applications for surface modification techniques.

2.1 Recent Research Trends

In development of an advanced fuel system, fabrication of a protective layer on fuel claddings has drawn positive attention to improved safety of nuclear reactors with development of accident tolerant fuels (ATFs). Currently, several research groups in USA consider coatings on the existing Zr-alloy cladding with functional materials including SiC, FeCrAl or Fe-Cr [5]. In this regard, we have selected FeCrAl alloy as the protective layer, and fabricated test samples using a sputtering system.

Unlike the small scale application, the large scale application essentially requires an efficient and stable modification technique for a large area. The coating dimension by the sputtering technique is dependent of a vacuum chamber size while a container size determines the coating dimension in the LbL method. Despite few relevant studies on boiling heat transfer, recent experimental works conducted at MIT implies that the LbL approach can be utilized for the large area modification [6]. Thus, for the large scale applications, we have employed another practical way, the LbL deposition technique. Moreover, brief explanations of two modification techniques and deposition mechanisms will be discussed in the following sections.

2.2 Sputtering Technique

FeCrAl layers can be fabricated using the sputtering system. Figure 2 shows a schematic image of the system is presented. The vacuum chamber contains target and substrate stages. Argon (Ar) gas is used as a working gas, and it flows through the chamber with a specific flowrate. A FeCrAl target is positioned at the upper part of the chamber. A bare SS316 substrate was placed on the substrate stage in the lower part of the chamber.

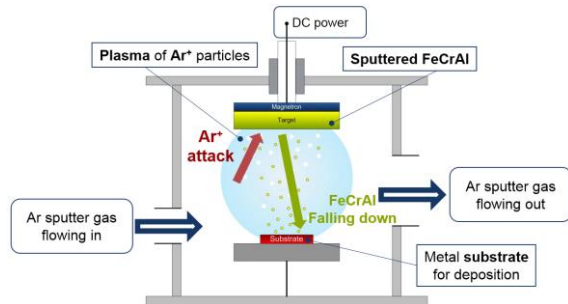


Fig. 2. Schematic of the sputtering system with Ar sputter gas

During the sputtering process, Ar ions (Ar^+) excited by a voltage sputter the target surface, and the target is bombarded with the energetic Ar^+ . As a result, sputtered FeCrAl atoms from the target surface are generated and experience a sequence of collisions with the existing particles in the chamber. After a series of scattering interactions, the atoms eventually arrive at the substrate surface, which results in a final stable FeCrAl layer [7].

2.3 Layer-by-Layer Deposition Technique

LbL deposition is a fabrication method for uniform thin film layers on various material substrates. Figure 3 shows the repetitive alternate immersion of substrates into a positively charged and a negatively charged solution [8, 9]. The repeating process results in stable film layers along the original surfaces of the substrates.

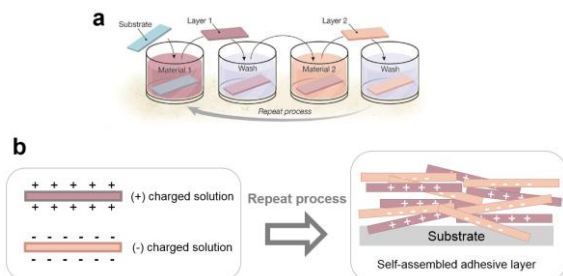


Fig. 3. Schematic of layer-by-layer assembly technology (a) LbL immersion process [8] (b) LbL-assembled thin film [9]

Compared to conventional coating methods, LbL deposition can be used on large areas and requires simple processing equipment because it is based on

solution-processing methods rather than high temperature processing [8]. The LbL method can be used to fabricate diverse coatings that are highly porous to form large surface area films of micro/nanostructured materials. In particular, the electrostatic force among bi-layers allows formation of more uniform, adhesive, and substantial coating layers than films depending on physical contacts to surfaces.

3. Sputtering System and Component Scale Application at THLAB

3.1 Experimental Assessment of Thermal Margin for FeCrAl-layered Heaters Fabricated by Sputtering Process

In order to assess the applicability of the modification technique, an assessment of thermal margin for FeCrAl-layered heaters fabricated by the sputtering system were carried out by measuring CHF. Pool boiling experiments were conducted with deionized water (DI water) at atmospheric pressure. Figure 4 shows CHF values obtained with substrate temperatures. As compared to the bare heater, all the heater samples exhibited enhancement in CHF ranged from 14% to 42%. It is evaluated that the CHF enhancement is attributed to increased roughness and improved rewetting to the hot dry spot.

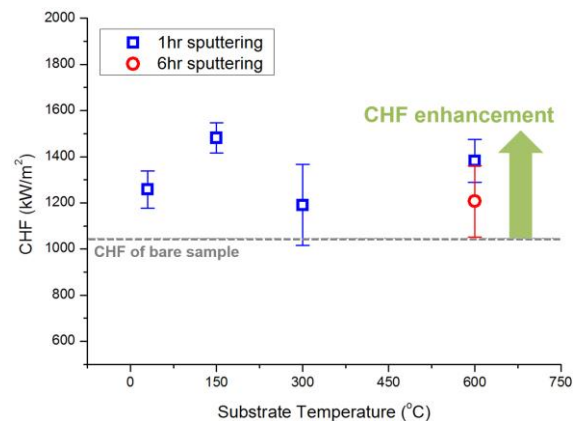


Fig. 4. Variation in CHF of the sputtering-layered heaters with substrate temperatures.

In addition to the CHF enhancement obtained from the pool boiling tests, a further investigation is currently in preparation. To employ the sputtering technique for the nuclear system, reactor-like environments or conditions is essentially considered. For this reason, an experimental facility developed will be described in the following discussion.

3.2 Description of Experimental Facility at THLAB

The typical operating conditions of primary cooling system for commercial light water reactors (LWRs) are known as ~15.5 MPa in pressure, 320 °C in the coolant

temperature, and $\sim 4.0 \text{ Mg/m}^2\text{-sec}$ in mass flux. In this environment, a sufficient amount of heat flux is required as high as $\sim 5.0 \text{ MW/m}^2$ for the CHF occurrence. To investigate boiling heat transfer and CHF phenomena at the reactor-like conditions, we have established the pressurized flow boiling heat transfer system. Employing scaling models suggested by Ahmad [10] and Katto [11], the R134a refrigerant was selected as the working fluid. With the R134a refrigerant, experimental conditions in the flow boiling system can be scaled down to $\sim 3 \text{ MPa}$ in pressure, $\sim 80 \text{ }^\circ\text{C}$ in the coolant temperature, and $\sim 0.4 \text{ MW/m}^2$ in heat flux. Figure 5 shows the developed experimental facility, which consists of the test section, heat removal components, circulation pump, and loop controller.



Fig. 5. Pressurized flow boiling loop at THLAB. Major design conditions with the R134a refrigerant are 3.5 MPa in pressure, 120 $^\circ\text{C}$ in temperature, 50 $^\circ\text{C}$ in inlet subcooling, and 2000 $\text{kg/m}^2\text{-sec}$ in mass flux.

Furthermore, to confirm the effects of sputtering conditions on bubble parameters and the CHF trigger mechanism, the test section is being modified to incorporate an annular channel and visualization system. After modification, optimized sputtering conditions and its impact on the performance of boiling heat transfer will be investigated. Consequently, we believe that the major findings from the investigation can contribute to the development of ATF.

4. Layer-by-Layer Deposition and System Scale Application at THLAB

4.1 Experimental Assessment of Thermal Margin for CNT-layered Heaters Fabricated by LbL Process

Similar to the experimental works discussed in Section 3, pool boiling tests were conducted with LbL-assembled heaters. In the LbL process, multi-walled carbon nanotubes (MWCNTs) and polyethylenimine

(PEI) were selected as the deposition materials. The test samples were prepared varying bi-layer numbers: 10, 20 and 40 bi-layer numbers. Figure 6 shows CHF values obtained. As compared to the bare heater, all the heater samples exhibited enhancement in CHF ranged from 39% to 107%. After the LbL process, the wettable, rougher and porous layer was formed on the bare surface, and the CHF enhancement was affected by the developed CNT layer.

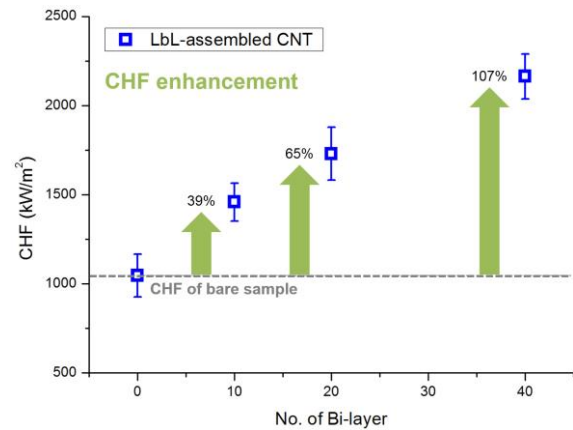


Fig. 6. Variation in CHF of the LbL-assembled heaters with bi-layer numbers.

4.2 Description of Experimental Facility at THLAB

As measures of severe accident mitigation, in-vessel corium retention (IVR) through external reactor vessel cooling (ERVC) and ex-vessel corium cooling system using an external core catcher has been proposed [12]. Several thermal-hydraulic features of the safety systems are considered such as the downward facing boiling heat transfer with low mass flow rate at low pressure.

To examine boiling behaviors occurring in the core catcher safety system, we have established the forced convective water boiling system with visualization equipment. The experimental facility as shown in Fig. 7 was designed to operate under the relatively low pressure condition up to 0.5 MPa. Moreover, circulation of water is controlled by a centrifugal pump with a range of mass flux from 40 to 400 $\text{kg/m}^2\text{-sec}$. The test section consists of the test section body, rectangular ducts, and copper block to supply heat flux. The test section body simulates the core catcher body, and was scaled down to have the rectangular channel with inclined by 10 $^\circ$ from the horizontal plane. The dimensions of the channel are 400 \times 131.5 \times 30 mm^3 in length, width and height, respectively. The test section is heated up indirectly through the copper block where an assembly of cartridge heaters with a nominal power of 4 kW/EA is installed. The active heating region of the test sample is 203 \times 95.5 mm^2 in length and width, respectively, which is contacted with the copper block.

Since many studs are installed to support the core catcher body, the flow of coolant and its streamlines can

be substantially distorted. It is important to assess how enough the thermal margin of the system with the relevant structures is. Thus, after evaluating CHF of the test system, a large area modification to the test section will be performed to enhance the thermal margin using the LbL deposition technique.

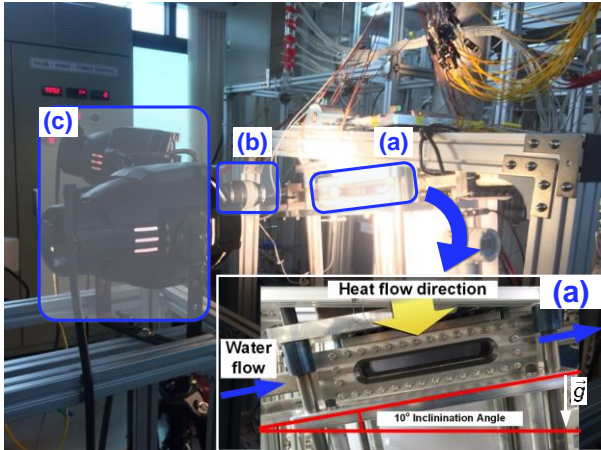


Fig. 7. Forced convective boiling loop with the high-speed video system. (a) test section, (b) high-speed camera, (c) light source.

5. Concluding Remarks

In this article, the cutting edge techniques for surface modification recently developed at THLAB were discussed: the sputtering and LbL deposition techniques. To assess the feasibility of the techniques, the pool boiling heat transfer experiments were carried out. For the applied boiling experiments, two distinct forced convective flow boiling systems have been established. The major findings and further directions at THLAB can be summarized as follows.

- The CHF enhancements were observed for for all the FeCrAl-layered heaters and LbL-assembled CNT heaters.
- Since each modification technique has the unique advantage respectively, the different applications are focused on according to the system scale.
- For the sputtering system, a protective layer can practically be deposited on a metal surface, and thus the technique can contribute to development of ATF.
- On the other hand, for the LbL deposition process, it has a significant strength in the large area modification. In this regard, the safety systems with the improved thermal margin could be achievable.
- For further investigations, forced convective flow boiling tests will be conducted under the applied boiling conditions that consider the reactor-like environments.

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