Pool Boiling Heat Transfer Characteristics of Chromium Coatings Deposited by RF Magnetron Sputtering

Gwang Hyeok Seo, Hong Hyun Son, Uiju Jeong, Gyoodong Jeun, Sung Joong Kim* Department of Nuclear Engineering, Hanyang University 222 Wangsimni-ro, Seongdong-gu, Seoul, 133-791, Republic of Korea *Corresponding author: sungjkim@hanyang.ac.kr

1. Introduction

The safety of nuclear power plants (NPPs) has been gradually developed over decades in the face of a few significant accidents. From the accident at Three Mile Island in 1980s, further evolutions for the nuclear safety have been carried out with analysis of system behavior with the probabilistic approach and development of passive safety systems. As a result, various types of NPPs currently adopt the advanced safety systems, which are AP1000 (USA), OPR1000 (South Korea), and EPR (EU) to mention a few. However, Since the Fukushima Dai-ich accident, a new challenge has been merged to require more sustainable and reliable evolutions even for an unexpected difficulty. Many researches have suggested fundamental changes to satisfy the safety requirements, including development of accident tolerant fuels (ATFs).

The adoption of coating techniques is one of promising approaches for ATF systems because surface modification with a highly oxidation-resistant material can prevent hydrogen generation and cladding embrittlement [1]. Compared to the development of a new cladding for the replacement of the current zirconium-based alloy cladding and new fuel forms instead of the current ceramic oxide fuels, the surface coating technique is cost-effective and easily applicable to the current LWR system with no significant design changes. Recently, a wide variety of oxidation-resistant materials have been proposed: iron-based alloys and SiC-based materials [2]. Among them, chromium (Cr) is suggested as a coating material for fuel claddings because it is known for has oxidation-resistant characteristic.

In order to assess the feasibility of coating techniques with an oxidation-resistant material, in this study chromium (Cr) film was deposited on a metal substrate via a physical vapor deposition (PVD) process. After preparing test specimens, pool boiling heat transfer experiments were carried out to investigate the boiling performance of both cases. Moreover, during a test, visualization works were conducted for a phenomenological understanding.

2. Experimental

Chromium (Cr) coated heaters are produced via a RF magnetron sputtering process, which is well known for a kind of PVD methods. After sophisticated

measurements for the surface characterization, a pool boiling heat transfer test is carried out.

2.1 Coating Process using RF Magnetron Sputtering

Cr film was deposited using a RF magnetron sputtering system. Figure 1 shows a schematic of the sputtering system. The metal or ceramic target is placed in a downward direction in the upper region of the vacuum chamber, and a substrate facing the target is positioned at the bottom region. The substrate and the target are 100 mm apart from each other. The size of the targets is 101.6 x 6.35 mm² in diameter and thickness, respectively. Purity of the Cr target is 99.95% (3N5). In the system, the RF power supply provides up to 1000W, and the substrate stage is heated to a maximum temperature of 600 °C.



Fig. 1. Sputter deposition system

Once a sputtering process begins, the chamber is evacuated to a base pressure of 2×10^{-3} Pa. Argon (Ar) sputter gas flows to the chamber, and the chamber pressure is maintained at a specific condition. Before starting deposition, the target is sputtered by Ar ions for 5 min to remove any contaminants on the surface. After the surface cleaning, the target is bombarded with the energetic Ar ions to generate sputter atoms from the target surface as described schematically in Fig. 2. During the PVD process, the sputtering conditions are carefully kept to produce a desirable film on the substrate, and the conditions are summarized in Table I.



Fig. 2. Physical sputtering process

Table I: Summary of Sputtering Conditions

Target	Cr
Flow rate, cc	30
Sputtering power, W	150
Sputtering time, hr	2
Pressure, Pa	0.13

2.2 Description of Experimental Apparatus and Heater Design

The experiments were conducted to investigate the effects of Cr film coated on a metal substrate in a pool of saturated DI water under atmospheric pressure. During a test, visualization work was also accompanied to observe the bubble behavior using high a speed camera system. Figure 3 shows a schematic of the pool boiling facilities used in this study.



Fig. 3. Pool boiling isothermal bed

The heater assembly consists of two copper blocks and a central heating section. Figure 4 shows a schematic of the test heater. The heating section is the coating substrate, and made of stainless steel grade 316 (SS316). The SS316 heater has dimensions of $42 \times 10 \times 2 \text{ mm}^3$ in length, width and thickness, respectively. Both ends of the heating section are bolted to each copper block, which transfers electrical power to the heater. After assembling, the side and back parts of the heater is insulated with epoxy. For the temperature measurement, a K-type TC is attached at the back of the heater. Voltage signals are collected at both ends of the heater to determine applied heat flux.



Fig. 4. Schematic of test heater assembly

3. Results and Discussion

3.1 Surface Characterization

The contact angle is an indicator used to quantify the surface wettability effect, which is a dominant parameter affecting the boiling heat transfer characteristics [3]. The Kandlikar's model suggests a contact angle is a major parameter, and the wettability effect on the CHF phenomenon [4]. Thus, a static contact angle measurement was performed to evaluate the wettability of the test heater. Figure 5 shows apparent contact angles on the heaters before and after the coating process. The resulting contact angles were estimated to be 72.7° for the bare sample and 16.4° for the Cr coated sample, respectively. The wettability of the Cr coated sample increased significantly, and a notable difference of 77% is evaluated.



Fig. 5. Apparent contact angles before and after coating on the SS316 substrate: 72.7° (up) and 16.4° (down)

In addition to the contact angle measurement, the surface morphology was investigated using SEM images.

Figure 6 shows plain and side views of Cr film deposited on the SS316 substrate. The Cr layer were uniformly and compactly developed on the substrate forming a nanoscale structure. As confirmed in the picture of side view, the coating layer consists a number of Cr pillars, a thickness of the layer is evaluated about $2 \mu m$.



Fig. 6. SEM images of Cr film: Plain view (left) and side view (right)

3.2 Critical Heat Flux of Cr Coated Heater

Figure 7 shows the resulting boiling curve and CHF value of about 1,100 kW/m² obtained in the experiment. The obtained CHF value were evaluated with existing models relevant to the heater conditions. The Zuber's model (1959) shown in Eq. (1) is widely adopted as a conventional CHF correlation, which was developed for an infinite flat heater [5,6]. The Zuber's prediction results in a CHF value of 1,107 kW/m², and this comparison indicates considerably similar values between the test and prediction.



Fig. 7. Boiling curve of Cr coated heater

$$q_{crit}^{"} = 0.13h_{fg}\rho_{v}^{1/2} \left[\sigma g\left(\rho_{l}-\rho_{v}\right)\right]^{1/4}$$
(1)

Although the surface characterization result indicates a great increase of the surface wettability, the Zuber's model does not reflect the wettability effect. The Kandlikar's model (2001) shown in Eq. (2) provides a CHF prediction incorporated the effects of the surface contact angle. The Eq. (2) shows that β and ϕ are the surface contact angle and heater orientation angle [4]. As the contact angle is 16.4° discussed in Section 3.1, the Kandlikar's prediction results in a CHF value of $1,528 \text{ kW/m^2}$. This prediction implies the wettability effect with the increased CHF value as many studies report. However, even the surface wettability increased considerably, the test result is in agreement with the Zuber's prediction, rather than the Kandlikar's.

$$\dot{q_{crit}} = h_{fg} \rho_v^{1/2} \left(\frac{1 + \cos \beta}{16} \right) \left[\frac{2}{\pi} + \frac{\pi}{4} (1 + \cos \beta) \cos \phi \right]^{1/2}$$
(2)

$$\times \left[\sigma g \left(\rho_l - \rho_v \right) \right]^{1/4}$$

O'Hanley et al. studied the effects of surface parameters, which are roughness, wettability, and porosity, and concluded that the wettability effect alone does not affect CHF. A dramatic increase of CHF was achieved on a wettable surface with the porous layer. In other words, the wettability effect appears with a combination of wettability and porosity [7]. The SEM images in Fig. 6 shows the Cr pillars piled up densely and uniformly, which indicates no porous structure. Thus, even a significant increase of wettability was obtained after the coating process, no notable change on CHF is expected.

4. Summary and Conclusions

In this study, Cr deposition on the SS316 surface was conducted using the sputtering process. Specifically, sophisticated surface characterization was performed with the wettability measurement and surface morphology analysis. Furthermore, the pool boiling heat transfer experiments were carried out to obtain the CHF value of the test heater. The major findings observed from this study can be summarized as follows.

• The surface wettability increased 77% after the sputtering deposition.

• However, despite the wettability increase, a similar value of CHF with the Zuber's prediction was obtained, which is about $1,100 \text{ kW/m}^2$.

• As one of oxidation-resistant materials, the CHF value on Cr coated heater implies that, adopting a Cr coated cladding may assure no hindrance in a steady-state operation in terms of CHF margin if accident tolerance of Cr coated claddings is confirmed in the future.

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REFERENCES

 S. J. Zinkle, K. A. Terrani, J. C. Gehin, L. J. Ott and L. L. Snead, Accident Tolerant Fuels for Lwrs: A Perspective, Journal of Nuclear Materials, Vol. 448, pp. 374, 2014.
K. A. Terrani, S. J. Zinkle and L. L. Snead, Advanced Oxidation-Resistant Iron-Based Alloys for Lwr Fuel Cladding, Journal of Nuclear Materials, Vol. 448, pp. 420, 2014. [3] S.J. Kim, I.C. Bang, J. Buongiorno, L.W. Hu, Surface wettability change during pool boiling of nanofluids and its effect on critical heat flux, International Journal of Heat and Mass Transfer, Vol. 50, pp. 4105-4116, 2007.

[4] S. G. Kandlikar, A Theoretical Model to Predict Pool Boiling CHF Incorporating Effects of Contact Angle and Orientation, Journal of Heat Transfer, Vol. 123, pp. 1071-1079, 2001.

[5] L. S. Tong and Y. S. Tang, Boiling Heat Transfer and Two-Phase Flow, 2nd Ed., Taylor and Francis, 1997.

[6] J. G. Collier and J. R. Thome, Convective Boiling and Condensation, 3rd Ed., Oxford Science Publication, 1994.

[7] H. O'Hanley, C. Coyle, J. Buongiorno, T. McKrell, L.-W. Hu, M. Rubner, and R. Cohen, Separate effects of surface roughness, wettability, and porosity on the boiling critical heat flux. Applied Physics Letters, Vol. 103, p. 024102, 2013.