

Critical heat flux enhancement of nuclear fuel cladding with a nano-porous oxide film by anodization

Hyoung Suk Yu^a, Yong Hoon Jeong^{a*}

^aDepartment of Nuclear and Quantum Engineering, Korea Advanced Institute of Science and Technology
291 Daehak-Ro, Yuseong-Gu, Daejeon 305-701, Republic of Korea

*Corresponding author: jeongyh@kaist.ac.kr

1. Introduction

The study and interest in severe accidents at nuclear power plants have been going on for a long time in a variety of ways. In particular, research on hydrogen explosion has been concentrated after the Fukushima accident [1, 2]. Among them, the accident tolerant fuel cladding reduces the oxidation of the steam and metallic materials, especially zircaloy cladding, at higher temperatures, thus fundamentally prevents the generation of hydrogen which causes hydrogen explosion in the Fukushima accident [3]. Therefore, it also increases the safety of the plant in addition to the removal equipment that eliminates hydrogen. Previous studies like ceramic cladding had problems applying the technology due to high prices and compatibility with the cladding. In this study, the critical heat flux of a cladding, which has with oxidation resistance using anodization method, was measured by anodization. Plus, to simulate conditions similar to those of nuclear power plants, the influence of anodic oxide films was confirmed under the condition of adding boron water.

2. Methods and Results

A pool boiling device was installed to assess the critical heat flux of the Zirlo plate. The following subsections explain each part of the test pool.

2.1 Test pool

The pool is made of polycarbonate, which has a size of 25cm x 25cm x 25cm (Width x Depth x Height). On the top of the test pool, there is a hole to discharge the internal steam to maintain the atmospheric pressure condition during the experiment. Also, the condenser is attached, so the steam is condensed, sent back to the test pool to maintain the concentration of the water. The pool's diagram is shown in figure 1 below.

2.2 Experimental Procedure

The experiments were carried out on specimens of 5, 10, 15, and 20 minutes, respectively, from non-anodized specimens. Zircaloy plate (Zr-Nb-Sn alloy, KEPCO Nuclear Fuel Company, Ltd., 0.8 mm thick) also known as ZIRLO was used in the experiment with anodization and CHF. Before anodization, the boiling side of Zirlo was polished with #600 and #1200 grit sandpaper to minimize the effect of surface roughness. The specimen was cleaned with alcohol and deionized water (DI) and dried by an air gun. The anodization was done in organic electrolyte made with ethylene glycol (95% purity,

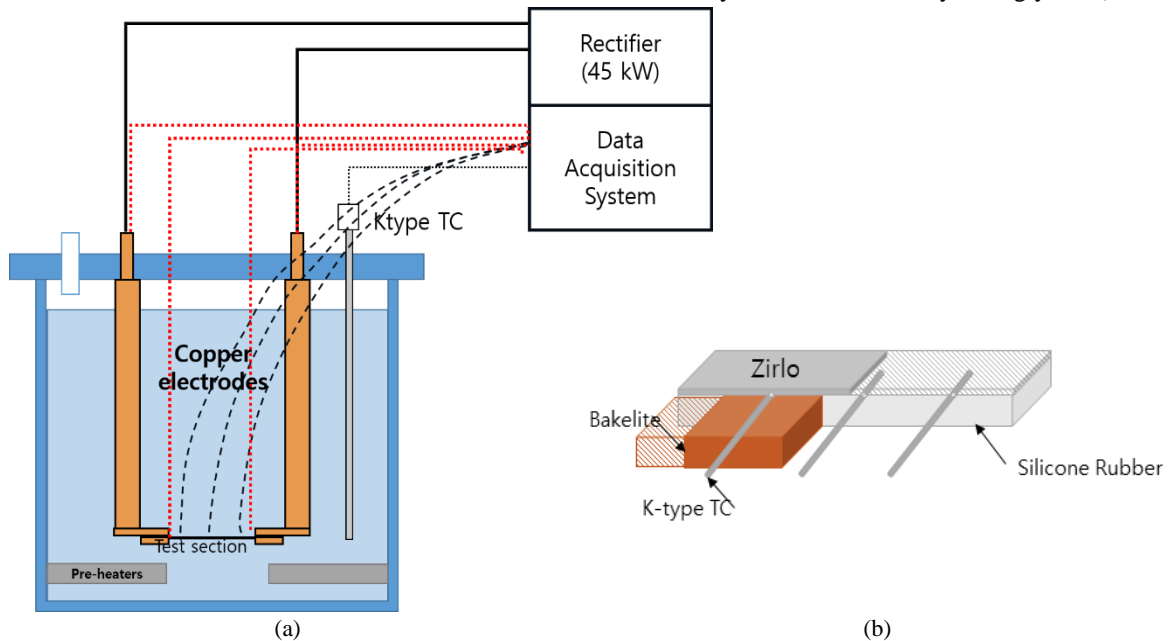


Fig. 1. The schematic diagram of (a) pool boiling apparatus and (b) holder.

Junsei) containing ammonium fluoride (NH₄F, Sigma-Aldrich Corporation, St. Louis, MO, USA) and DI water. During anodization heat generated therefore electrolyte was cooled by alcohol at 0°C. The cathode was parallel to specimen 5cm apart, and the applied voltage was fixed 90V.

The pool boiling experiments were conducted under atmospheric condition. The temperature in the pool was kept saturated with a pre-heater located below of the pool, and the heating method of the specimen was Joule heating (direct heating) using a DC rectifier (15V, 3000A, 45kW). The bottom of the sample was insulated using silicone rubber and bakelite. So, heat is only transferred through the top and sides of the specimen. The temperature of the specimen is measured with three K-type thermocouples located below the sample and recorded via DAS (Agilent 34980A) along with applied current and voltage. The critical heat flux (CHF) point is the point at which the temperature rises rapidly, and its value is calculated using the formula follow :

$$q_{CHF} = \frac{V_{CHF} \times I_{CHF}}{A_h} \quad (1)$$

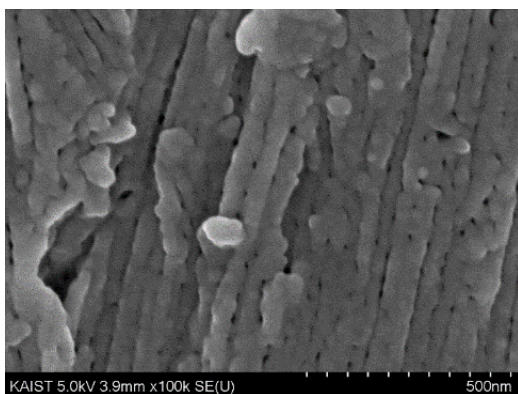
where q_{CHF} is the CHF of the specimen; V_{CHF} is the voltage of specimen; I_{CHF} is the current of the specimen; A_h is the area of heat is transferred.

2.3 Contact angle measurement

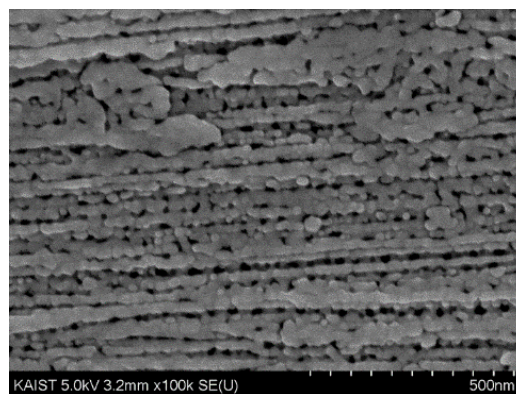
The contact angles were measured for each test specimen before the experiment. The specimen was cleaned before measuring the contact angle. The contact measurements were done with 5 μ L of the DI water at least 5 different points of the specimen.

2.4 Result

The results of increased critical heat flux after anodization are shown in figure 2. The experiment was conducted more than 5 times for each condition and at the identical time more than 20 minutes to minimize the difference due to boiling time.



(a)



(b)

Fig. 3. SEM images (magnification x100k) (a) the 5min anodized Zirlo, (b) 20min anodized Zirlo

In each condition, the critical heat flux of bare specimen is 819 kW/m², 5 minutes is 1004 kW/m², 10 minutes is 1292 kW/m², 15 minutes is 1235 kW/m², and last 20 minutes is 1295 kW/m². The percentages of enhancement are 23%, 58%, 51%, and 58%, respectively. The rate of critical heat flux enhancement rises rapidly to 10 minutes of anodization and then converges without further enhancement. This result is shown in the figure 3 SEM image, where the nano-porous surface morphology was not fully formed in the 5 minutes anodized surface. However, after 10 minutes of anodization, we can see the clear nano-porous surface.

Measured contact angle data and a picture are shown in the Fig.4 below. The contact angle was measured as 50.8° from the bare specimen, 38.4° from the 5 minutes, 37.9° from the 10 minutes, 31.2° from the 15 minutes, and 28.5° from the 20 minutes. The increased in wettability also shows a tendency similar to the increase in critical heat flux.

The results of the experiment under the boric acid condition are shown Fig.5 below. For the bare specimen, the critical heat flux was rising under boric acid condition. Meanwhile, the anodized sample did not show a significant change in boric acid condition. In the bare specimens, critical heat flux was significantly

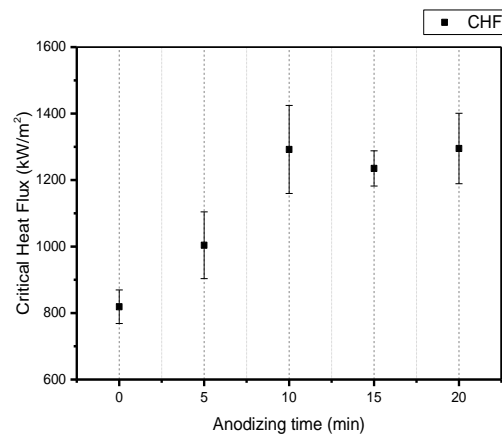


Fig. 2. The CHF value under DI water condition

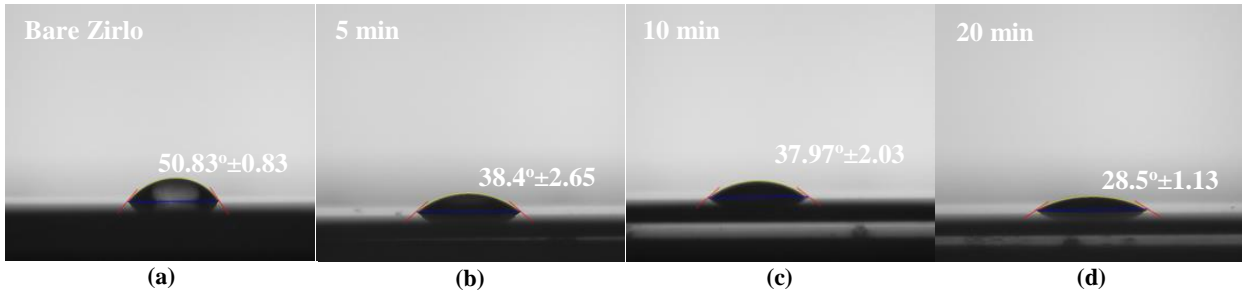


Fig. 4. Contact angle image of (a) bare Zirlo, (b) 5min, (c) 10min, (d) 20min anodized Zirlo

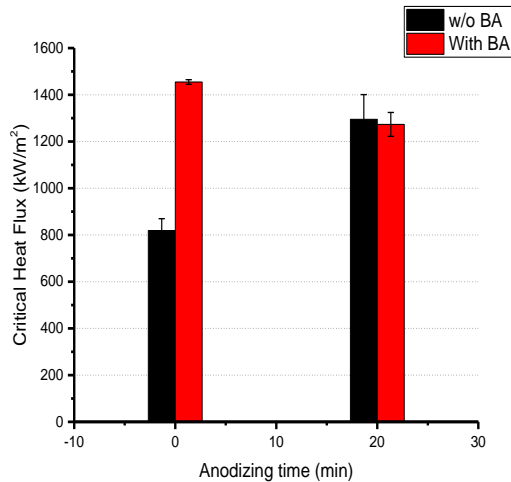


Fig. 5. CHF value of bare Zirlo and 20 min anodized Zirlo with or w/o boric acid condition.

increased to about 40% under boric acid condition. There was no change in the case of the oxide film on it

3. Conclusions

In this study, the improvement of the critical heat flow rates of the anodized zirlo specimen was confirmed through a pool boiling experiment. Besides, the pool boiling experiments were conducted under the condition of boron to verify an actual reactor water chemistry condition. The wettability of surface rises after the anodization. Moreover, the critical heat flux also rises as wettability rises. At this time, the increase in the critical heat flux is saturated after 10 minutes of anodization. Furthermore, under the boric acid condition, the anodized specimen showed no significant difference compared to the bare specimen. This is because the pre-formed oxide layer prevented the oxidation by contact with boric acid water.

REFERENCES

- [1] Zinkle, Steven J., et al. Accident tolerant fuels for LWRs: A perspective, *Journal of Nuclear Materials*, Vol. 448, No. 1, pp. 374-379, 2014.
- [2] Cronenberg, A. W. In-vessel Zircaloy oxidation/hydrogen generation behavior during severe accidents. No. NUREG/CR-5597. Nuclear Regulatory Commission, Washington, DC (USA). Div. of Systems Research; Engineering Science and Analysis, Albuquerque, NM (USA), 1990.

[3] Hirose, Katsumi., 2011 Fukushima Dai-ichi nuclear power plant accident: summary of regional radioactive deposition monitoring results, *Journal of environmental radioactivity*, Vol. 111, pp. 13-17, 2012.