Multi-Metallic Layered Tubing Fabrication for ATF Cladding Application

Joonho Moon [a], Sungyu Kim [a], Ji Hyun Kim [b], Michael P. Short [c], Chi Bum Bahn [a*]
[a]*School of Mechanical Engineering, Pusan National University, 2, Busandaehak-ro 63beon-gil, Geumjeong-gu, Busan, 46241*
[b]*Department of Nuclear Science and Engineering, School of Mechanical and Nuclear Engineering, Ulsan National Institute of Science and Technology, 50, UNIST-gil, Eonyang-eup, Ulji-gun, Ulsan 44919,*
[c]*Department of Nuclear Science and Engineering, Massachusetts Institute of Technology, 77 Massachusetts Ave., Room 24-204, Cambridge, MA 02139*
**Corresponding author: bahn@pusan.ac.kr*

1. Introduction

After the Fukushima accident, in order to prevent or mitigate the severe accident, many accident-tolerant fuel (ATF) cladding materials have been studied in the world, such as coated Zr alloy, FeCrAl, SiC, Cr-Mo alloy etc. [1-2]. Multi-metallic layered composite (MMLC) cladding is one of the potential ATF cladding designs. Under the lead-bismuth eutectic (LBE) environment, MMLC fuel cladding has been shown to have good properties [3]. In order to apply this concept to light water reactors (LWRs), we have redesigned the MMLC cladding concept. It is mainly composed of Zr alloy, FeCrSi alloy, and buffer materials in between them. In the fabrication process of MMLC fuel cladding, the layers are bonded onto the Zr billet, not the Zr cladding. After bonding process, extrusion and pilgering process are conducted in order to decrease MMLC mother tube thickness and size. Currently, the fabrication of MMLC fuel cladding has been completed until the bonding process. In this paper, we report the progress and lessons learned in MMLC fuel cladding development for LWR.

2. Background and Design

In the MMLC fuel cladding, the inside layer is composed of zirconium alloy that has low neutron absorption cross-section and high neutron irradiation resistance. The outside layer is composed of FeCrSi alloy, which has high resistance to steam oxidation. However, Zr and Fe, the main elements of MMLC fuel cladding, form the intermetallic compound in the high temperature. Therefore, diffusion barrier layers are required between Zr alloy and FeCrSi alloy layers in order to prevent the formation of intermetallic compound. The Zr-Fe phase diagram is shown in Figure 1.

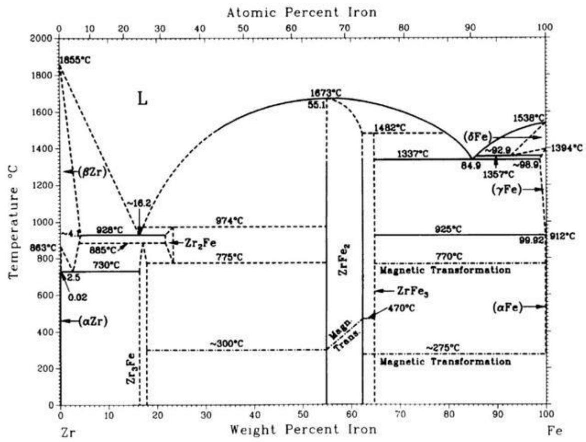

Fig. 1. Zr-Fe phase diagram [4]

Figure 2 shows the changes in MMLC fuel cladding design. Based on phase diagrams, Cr or Nb element does not form any intermetallic compound with both Zr and Fe. Therefore, we selected these elements as the buffer layers at the first time. However, Nb did not have diffusion bond to Cr well, leading to weak adhesion. Considering this, the Nb layer was switched to a Ti layer. However, Ti and Cr can form an intermetallic compound based on the Ti-Cr phase diagram. Thus, a Mo interlayer was added to prevent this intermetallic compound formation. The buffer layers of Ti, Cr, Mo were expected to prevent the intermetallic compound formation. However, the fabrication process of Cr-Mo layer was very difficult, it was raised that Cr layer can cause cracking during pilgering process because of brittleness of Cr. For this reason, Cr was replaced by V-Cr-Ti alloy. V-Cr-Ti alloy does not form intermetallic compound with Fe. Furthermore, this V alloy was reported to have successful bonding with steels [5]. Therefore, we finally selected Ti and V-Cr-Ti alloy as the buffer layer.

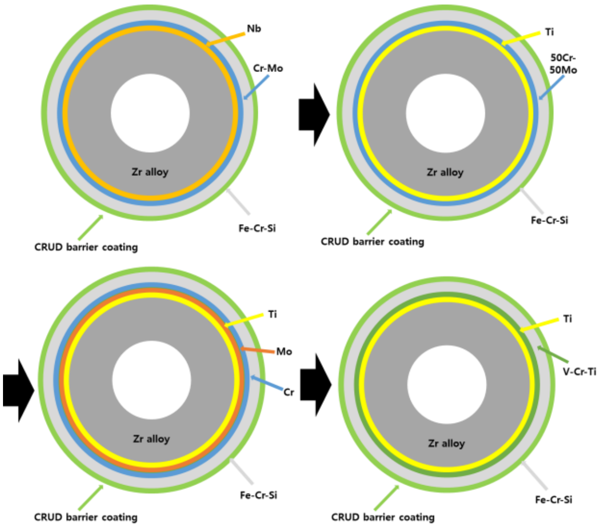

Fig 2. Changes in MMLC fuel cladding design

3. Fabrication & Evaluation of HIP Specimens

Before fabrication of the MMLC fuel cladding mother tube, in order to check any potential process problems, small plate specimens were fabricated. Two types of specimens were fabricated: 3-layer and 4-layer. 3-layer specimen is consisted Zr alloy, Ti and Fe12Cr2Si, whereas 4-layer specimen is consisted Zr alloy, Ti, V and Fe12Cr2Si. Because the zircaloy-4 and V-Cr-Ti alloy of MMLC fuel cladding design were difficult to obtain, small plate specimens were fabricated using Zr 705 commercial alloy and pure V instead. Figure 3 shows the schematic of small plate specimens.

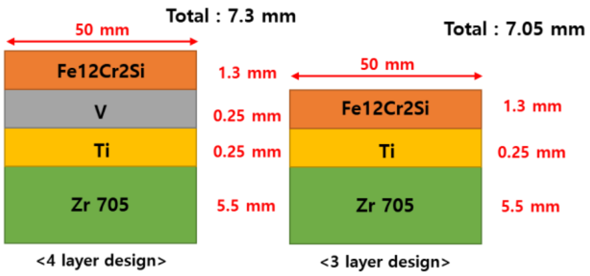

Fig. 3. Schematic of small plate specimens

3.1. Fabrication and cold rolling

In order to bond materials, hot isostatic pressing (HIP) process and heat treatment were used. After bonding process, cold rolling was conducted in order to simulate the pilgering process of MMLC mother tube. The total process of small plate specimens is shown in Figure 4.

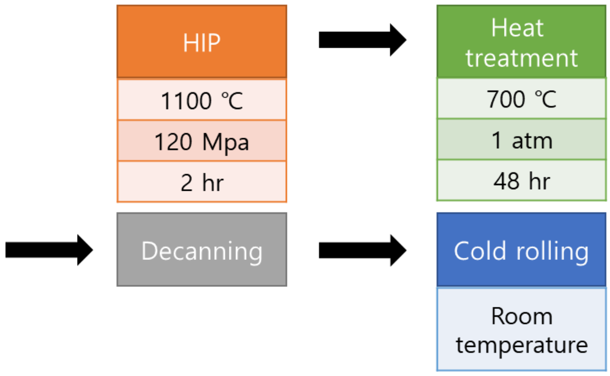

Fig. 4. Total process of small plate specimens

During conducting the HIP process and heat treatment, these specimens were packed in the stainless steel case in order to prevent oxidation and increase bonding force. After HIP and heat treatment process, it was unpacked. The cold rolling process was continuously conducted until cracks occurred in the specimens.

3.2. Evaluation results

In the case of 3-layer specimen, outermost layer was broken when cold rolling it up to 1.6% thickness reduction. On the other hand, in the case of 4-layer specimen, the specimen shape was maintained when cold rolling it up to 22% thickness reduction. In order to check the chemical composition of layers, the SEM-EDS analysis of each cross-section was conducted. Figure 5 shows the overall shape and each layer's chemical composition of two specimens after the cold rolling process.

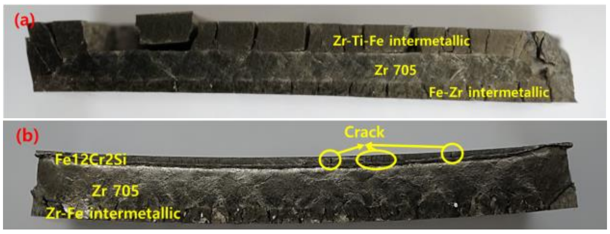

Fig. 5. Overall shape and each layer's chemical composition of small plate specimens after cold rolling: (a) 3-layer, (b) 4-layer

As shown in Figure 5, the top layer of 3-layer small plate specimen is a Zr-Ti-Fe intermetallic compound. This result indicates that the Ti single layer cannot be an effective diffusion barrier. Also, the bottom Zr-Fe intermetallic compound is formed because of the contact between Zr layer and stainless steel canning materials. In the 4-layer small plate specimen, Fe12Cr2Si layer is still remained, and Zr-Fe intermetallic compound is not formed. It shows that the dual layers of Ti and V can prevent the formation of Zr-Fe intermetallic compound. Moreover, although some cracks occurred in Fe12CrSi layer after cold rolling, the overall specimen shape is maintained. Therefore, the pilgering process is expected to be conducted well under suitable heat treatment conditions.

4. Fabrication & Evaluation of Mother Tube

In the small plate specimen result, the Ti single layer could not prevent Zr-Fe intermetallic compound formation. However, if the Ti single layer is thick enough, it may be able to prevent the intermetallic compound formation. Also, the fewer the layers, the better the pilgering process. Therefore, two types of MMLC fuel cladding mother tube were fabricated: 3-layer and 4-layer. 3-layer mother tube has only the Ti single barrier layer, 4-layer mother tube has Ti and V-Cr-Ti dual barrier layers. The bonding process of MMLC mother tubes were conducted through the international collaboration.

4.1. 3-layer MMLC mother tube

In order to bond the materials, Zircaloy-4, Ti, Fe12Cr2Si alloy were prepared. Fe12Cr2Si alloy was made using overlay welding process on a carbon steel pipe. After building up the Fe12Cr2Si weld deposit layer, the carbon steel pipe was removed. Each material was bonded using a shrink-fit process. Figure 6 shows the fabrication process and photographs of 3-layer mother tube.

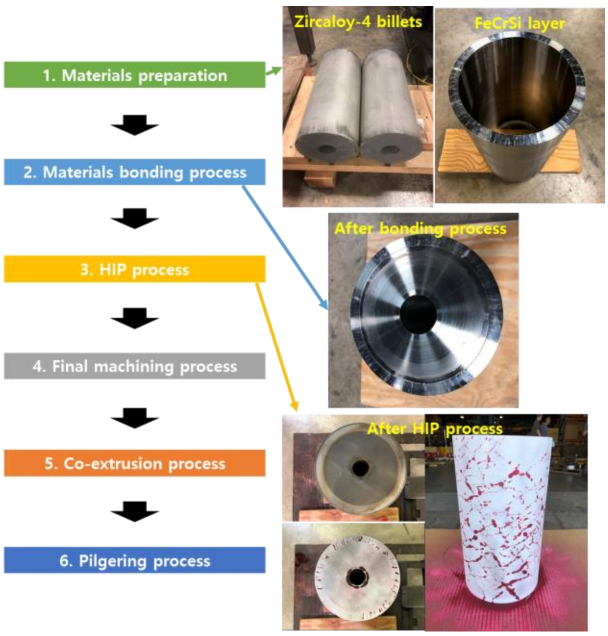

Fig. 6. Fabrication process and picture of 3-layer mother tube

After the HIP process, it is observed that many cracks occurred in Fe12Cr2Si layer. In order to check the cause of cracking, the SEM-EDS analysis of cross-section was conducted. The EDS mapping result of 3-layer MMLC mother tube is shown in Figure 7.

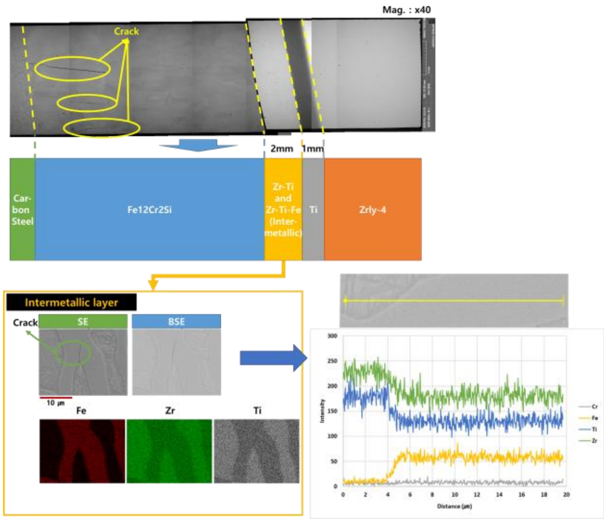

Fig. 7. EDS mapping and line-scan results on the cross-sectional images of 3-layer mother tube after HIP process

Figure 7 shows some cracks in Fe12Cr2Si layer and forming of Zr-Ti-Fe intermetallic compound. This result shows that a thick Ti single layer cannot prevent the formation of Zr-Fe intermetallic compound. It is the same result as the HIP specimen. It is also observed that the intermetallic layer actually consists of two different intermetallics: Zr-Ti and Zr-Ti-Fe, which seem to be separated in the intermetallic layer. Some small cracks are also observed in this layer. However, it appears that the cracks of Fe12Cr2Si layer and intermetallic layer is not connected to each other, suggesting that two cracking phenomena occurred independently. The reason for the occurrence of some cracks in Fe12Cr2Si layer is under evaluation.

4.2. 4-layer MMLC mother tube

In the 3-layer MMLC mother tube fabrication result, many cracks were observed after HIP process. Therefore, when making the 4-layer MMLC mother tube, the HIP process was removed. Other process is the same as the 3-layer MMLC mother tube. The 4-layer MMLC mother tube consists of zircaloy-4, Ti, V13Cr5Ti and Fe12Cr2Si alloy. Figure 8 shows the fabrication process and the assembled 4-layer mother tube.

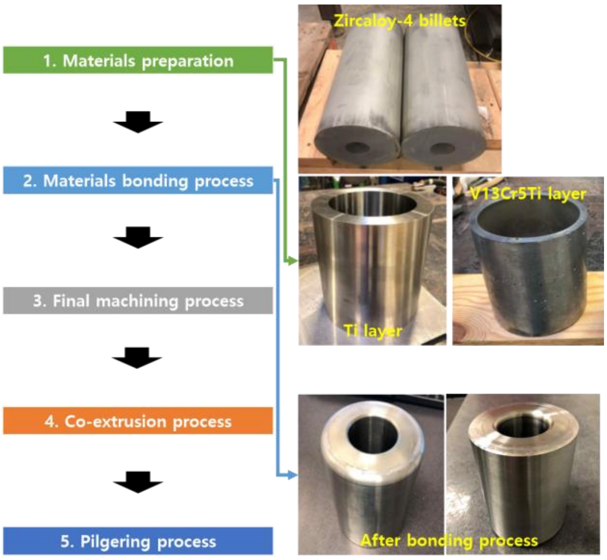

Fig. 8. Fabrication process and photographs of 4-layer mother tube

Currently, the fabrication of 4-layer mother tube was proceeded until the final machining process. It is planned to conduct the extrusion process and pilgering process in order to decrease the thickness and size of MMLC mother tube until the size of commercial Zr fuel cladding.

5. Conclusions and Future Work

The layer-by-layer design of MMLC mother tube evolved to compensate for the weakness during the fabrication trial. Cold rolling was conducted with small plate specimens to identify layers that could be problematic during the pilgering process of MMLC mother tube. The small plate specimen results showed that the 3-layer specimen containing only Ti single barrier layer cannot prevent the formation of Zr-Fe intermetallic compound, whereas the 4-layer specimen containing V and Ti dual barrier layer can prevent the formation of Zr-Fe intermetallic compound. Based on these results, 3-layer and 4-layer MMLC mother tube were prepared. It was confirmed that 3-layer MMLC mother tube containing a thick Ti single barrier layer cannot prevent intermetallic compound formation, which is the same result as small specimens. In addition, after the HIP process, cracks occurred in the Fe12Cr2Si layer, the outermost layer of the 3-layer MMLC mother tube. Currently, the 4-layer MMLC mother tube has been completed until the bonding process. It is planned to conduct the extrusion and pilgering process in order to decrease the thickness and size of MMLC mother tube until the size of commercial Zr fuel cladding.

ACKNOWLEDGEMENT

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REFERENCES

[1] H. G. Kim, J. H. Yang, W. J. Kim, and Y. H. Koo, Development status of accident-tolerant fuel for light water reactors in Korea, Nuclear Engineering and Technology, Vol.48, p.1, 2016.
[2] K. A. Terrani, Accident tolerant fuel cladding development: Promise, status, and challenges, Journal of Nuclear Materials, Vol.501, p.13, 2018.
[3] M. P. Short, R. G. Ballinger, and H. E. Hänninen, Corrosion resistance of alloys F91 and Fe–12Cr–2Si in lead–bismuth eutectic up to 715 C, Journal of Nuclear Materials, Vol.434. p.259, 2013.
[4] D. Arias, and J. P. Abriata, The Fe− Zr (Iron-Zirconium) system, Bulletin of Alloy Phase Diagrams, Vol.9, p.597, 1988.
[5] J. M. Chen, V. M. Chernov, R. J. Kurtz, and T. Muroga, Overview of the vanadium alloy researches for fusion reactors, Journal of Nuclear Materials, Vol.417, p.289, 2011.