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The effects of Cr^{2+} ion irradiation on the formation of precipitation in multi-metallic layer composite cladding processed by hot isostatic pressing

Tae Yong Kim, Jeong Hyeon Lee, Ji Hyun Kim [*]
Department of Nuclear Engineering, School of Mechanical, Aerospace and Nuclear Engineering,
Ulsan National Institute of Science and Technology, 50 UNIST-gil, Ulsan 44919
[*]*Corresponding author: kimjh@unist.ac.kr*

1. Introduction

In light water reactor(LWR), zirconium alloys which have low thermal neutron absorption cross section have been used broadly. However, when Loss Of Coolant Accident(LOCA) at nuclear power plant is occurred such as the Fukushima Daiichi melt down in 2011, the zirconium cladding reacts with high temperature steam and produces vast amounts of hydrogen, which can lead to explosions and the release of radioactive materials into the environment. To solve this problem, a cladding material which has high oxidation resistivity and low hydrogen generation is required although fuel cladding temperature are rising at LOCA.

In order to avoid these problems in LWRs going forward, we aim to develop a multi-metallic layered composite (MMLC) which compromises between the neutronic advantages of zirconium alloys and the accident-tolerance. A schematic diagram of our MMLC cladding concept is shown in Fig. 1.

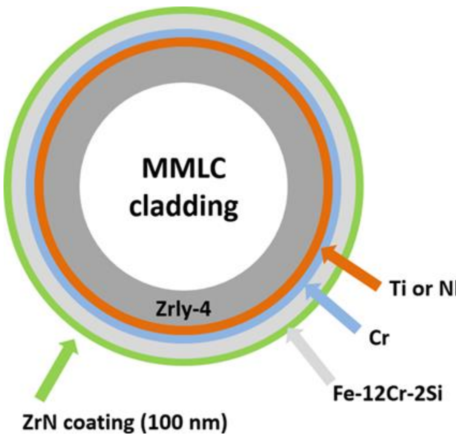

Fig. 1. Schematic diagram of cross-section of MMLC fuel cladding concept.

The layers of the proposed MMLC are designed to perform specific functions unattainable by single alloys. Fuel cladding composed of a iron-based alloy, overlaid onto a Zircaloy base, could reduce the amount of Zr in the reactor, resulting in less hydrogen evolution during a severe accident. In addition, a MMLC cladding will be less susceptible to sudden, brittle failure of fuel cladding due to directional hydride formation in Zircaloys. Finally, the water-facing layer of Fe-12Cr-2Si layer will resist severe accident corrosion better than Zircaloys. Diffusion barrier layers of Cr(or Cr+Mo) and Ti(or Nb) must be used in between Zr and an iron-based alloy, to avoid detrimental eutectic phase or intermetallic formation [1].

A completely new concept is required to investigate the irradiation effect on the MMLC fuel cladding materials. The irradiation evaluation is important to survey unrevealed effects of irradiation on multi-metallic layer. At manufacturing process of MMLC fuel cladding, there is welding process to bond each composite material like Fig. 2.

Fig. 2. Overlay-welding process to bond each composite material

However, it can make residual stress related to radiation induced segregation or diffusion [2]. In this situation, irradiation may contribute to formation of precipitation or brittleness near welding region. Therefore, in this research, our goal is focused on formation of precipitations at irradiation environment on MMLC fuel cladding.

In specific procedure, the microstructure including precipitation have been investigated by using transmission electron microscope (TEM).

2. Experimental

2.1 Alloys

There are two type test materials in this study. One is Zr-based alloy and the other is MMLC plate. And MMLC plate has four layers which composed of Fe-12Cr-2Si, Cr, Ti and Zircaloy-4 in order from outer layer. Chemical compositions of Zr-based alloy, Zircaloy-4 and Fe-12Cr-2Si are shown in Table. 1.The MMLC plate were fabricated via Hot Isostatic Pressing (HIP) process at 1000 °C and 15 ksi pressure for 4 hour. HIP process has been demonstrated to be an efficient way to produce laboratory-scale samples of diffusion bonded metallic composites [3]. A basic schematic of the HIP process we used is shown in Fig. 3. The laboratory-scale MMLC plates that have been produced have been aged at 700 °C for 4, 8, and 16 days in order to further enhance the metallurgical bonding between the metallic layers.

Table I: Chemical composition in wt. %

	Zr-alloy	Zircaloy-4	Fe-12Cr-2Si
Zr	Bal.	Bal.	-
Nb	1.02	-	-
Sn	0.96	1.2	-
Fe	0.094	0.18	85.5
Cr	0.0079	0.07	12.2
Si	0.0040	0.012	2.12
C	0.0060	0.027	0.007
N	0.0022	0.007	0.002

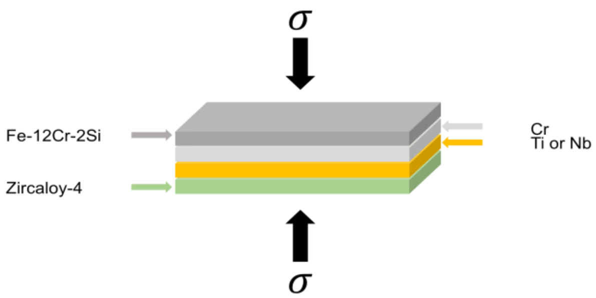

Fig. 3. Schematic of the HIP process used to diffusion bond of MMLC plate.

2.2 heavy ion irradiations

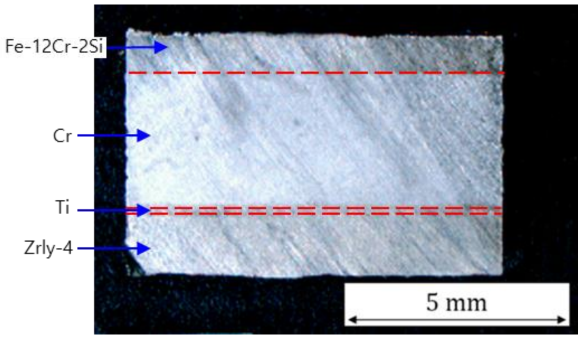

Fig. 4. MMLC plate for irradiation

Irradiation experiments were conducted using a Low Energy Ion Beam Facilities-Metal in the Korea Multi-purpose Accelerator Complex (KOMAC). The MMLC plate is in the form of 8 mm x 5 mm x 1 mm like Fig. 4. And the Zr-alloy plate size is 10 mm x 1 mm x 1 mm. Heavy ion irradiations were conducted using 140 keV Cr^{2+} at room temperature. Irradiated surface of MMLC plate is selected with including all composite layers. And samples were irradiated to about 30 dpa at the depth of 100 nm from the irradiated surface calculated using the SRIM code [4]. The dose rate was ~7x10^{-4} dpa s^{-1}.

2.3 Sample preparation and data analysis

Specimens for TEM analysis were prepared using the focused-ion-beam (FIB) system. Specially, the specimens for evaluation of MMLC plate were obtained from three welding regions such as Fe-12Cr-2Si/Cr, Cr/Ti, and Ti/Zircaloy-4. Thereby, 4 type specimens including Zr-alloy will be analyzed. However, this paper will deal with only Zr-alloy, welding region between Fe-12Cr-2Si/Cr and welding region between Cr/Ti. The final goal of this experiment is analysis of formation of precipitations. TEM analysis was conducted for compare un-irradiated and irradiated specimens.

3. Results

3.1 Analysis of Zr-alloy

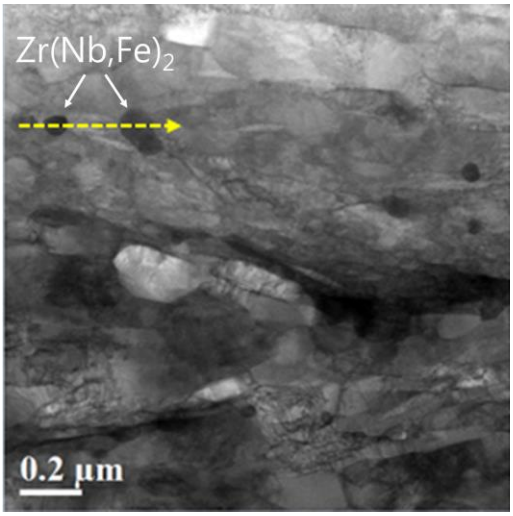

Fig. 5. TEM analysis of un-irradiated Zr-alloy.

Un-irradiated Zr-alloy specimen shows laves-phase precipitations at Fig. 5. Thorough line EDS analysis from Fig. 6 along the yellow line in Fig. 5, the two precipitation are composed of Zr, Nb and Fe. Size of the precipitations is about 100 nm. And Irradiated Zr-alloy was being analyzed. As soon as possible, behavior of that at irradiation environment will be evaluated. Through literature study, at irradiated Zr-alloy, the precipitations are expected to have smaller size and low number density. Moreover, Fe atoms included in the precipitation may be diffused into matrix by irradiation [5]. This is different from radiation induced segregation phenomena. High dose rate effects on annihilation to prevent segregation of elements into grainboundaries. Therefore, the precipitations may be shrink [6].

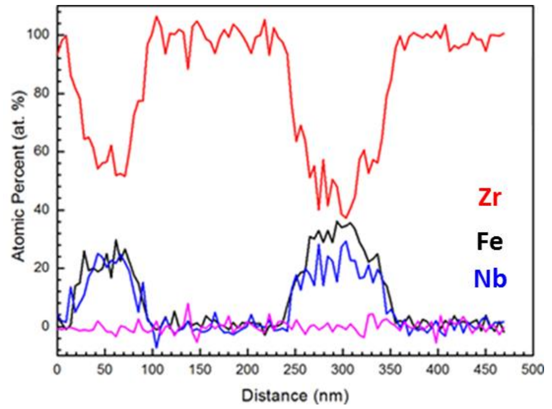

Fig. 6. EDS analysis of Zr(Nb,Fe)2 in un-irradiated Zr-alloy.

3.2 Analysis of welding region between Fe-12Cr-2Si/Cr

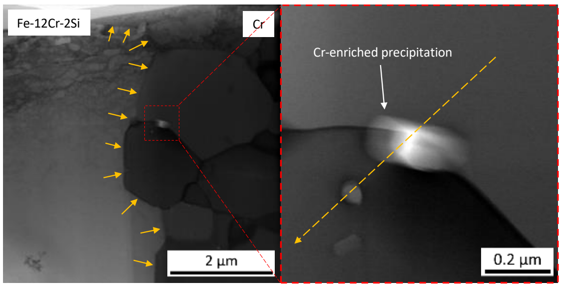

Fig. 7. STEM image of a thin foil of un-irradiated bonding region prepared using Dual-Beam FIB-SEM.

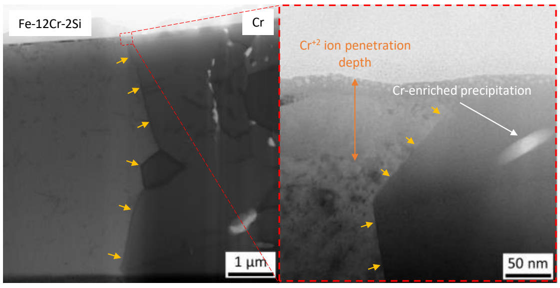

Fig. 8. STEM image of a thin foil of irradiated bonding region prepared using Dual-Beam FIB-SEM.

welding region of Fe-12Cr-2Si and Cr layer compare un-irradiated and irradiated specimen. At un-irraidated Fe-12Cr-2Si layer, there is no precipitation. On the other hand, at grain boundary of un-irradiated Cr layer, 200 nm size precipitation is formed. The precipitation has many chromium, 26 weight percent iron and 25 weight percent silicon. Next, Irradiated Fe-12Cr-2Si layer still has no precipitation. However, irradiated Cr layer shows a small amount of precipitations with 50 nm size. The reason is considered as precipitates is subjected to be amorphized at high dpa and elements composing the precipitates are diffused into surrounding grains [5].

3.3 Analysis of welding region between Cr/Ti

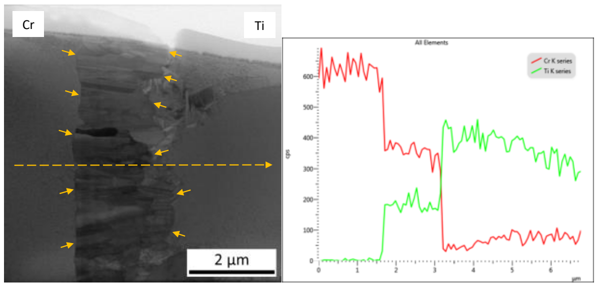

Fig. 9. STEM image and EDS analysis of a thin foil of un-irradiated Cr and Ti bonding region prepared using Dual-Beam FIB-SEM.

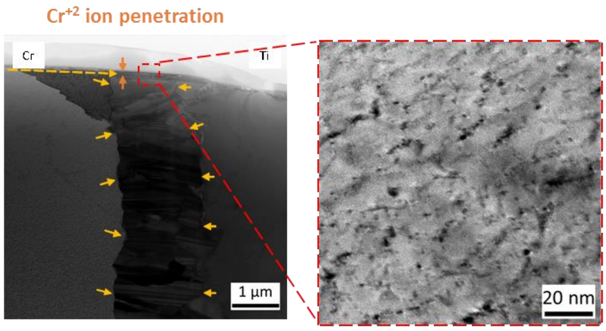

Fig. 10. STEM image and EDS analysis of a thin foil of irradiated Cr and Ti bonding region prepared using Dual-Beam FIB-SEM.

Last specimen which is welding region of Ti and Cr layer form no precipitation whether or not specimens are irradiated. Instead, there is 2 um thickness intermetallic phase, $TiCr_2$ between Cr and Ti layer. The intermetallic phase seems to be resulted from pre-heat treatment like HIP and aging process. And we can observe void and dislocation at irradiated intermetallic phase.

3. Conclusions

The research investigated irradiation effect on behavior of precipitation about multi-metallic layer composite plate. electron microscope analysis was conducted at welding regions between Fe-12Cr-2Si/Cr layer and between Cr/Ti layer, and Zr-alloy.

At un-irradiated Zr-alloy specimen, $Zr(Nb,Fe)_2$ laves-phase is observed. However, the precipitation is expected to be small and have low number density at irradiated environment with high dpa.

Welding region between Fe-12Cr-2Si/Cr layer shows precipitations composed of Cr, Fe and Si at Cr layer. But the specimen irradiated by Cr2+ ion has a few of small size precipitations.

Last specimen about welding region between Ti/Cr layer forms $TiCr_2$ intermetallic phase instead of forming precipitation. And irradiated welding region shows some void and dislocation.

ACKNOWLDEGEMENT

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REFERENCES

[1] M. Short, R. G. Ballinger, A Functionally Graded Composite for Service in High-Temperature Lead-and Lead-Bismuth-Cooled Nuclear Reactors-1: Design, Nuclear Technology, Vol.177, p. 366-381, 2012.

[2] Z. Jiao, and G. S. Was, Segregation behavior in proton- and heavy-ion-irradiated ferritic-martensitic alloys, Acta Materialia, Vol.59, p. 4467-4481, 2011.

[3] M. P. Short, S. Morton, S. E. Ferry, and R. G. Ballinger, Diffusional stability of ferritic-martensitic steel composite for service in advanced lead-bismuth cooled nuclear reactors. International Heat Treatment and Surface Engineering, International Heat treatment and Surface engineering, vol.4, p. 74-80, 2013.

[4] J. F. Ziegler et al., SRIM-The Stopping and range of Ions in Solids. 2009

[5] H.H. Shen et al., proton irradiation effects on the precipitate in a Zr-1.6Sn-0.6Nb-0.2Fe-0.1Cr alloy, Journal of Nuclear Materials, Vol.452, p. 335-342, 2014

[6] Friedrich Garzarolli et al., effect of high neutron fluences on microstructure and growth of zircaloy-4, ASTM, 1989.