

Development of Basic Technology for FeCrAl-based LWR Cladding

Sungyu Kim^{a*}, Joonho Moon^a, Chi Bum Bahn^{a**}

^a School of Mechanical Engineering, Pusan National University, Busan 46241, South Korea

*Presenting author: sgkim@pusan.ac.kr

**Corresponding author: bahn@pusan.ac.kr

1. Introduction

Zirconium alloy cladding has good corrosion resistance in the operating environment of pressurized water reactor and boiling water reactor, but rapid oxidation reaction occurs when the loss-of-coolant accident occurs as in the Fukushima nuclear power plant accident. If the hydrogen generated by this oxidation reaction accumulates inside the containment, hydrogen explosion may occur [1].

For this reason, the materials such as SiC, Mo and FeCrAl alloys, which are resistant to corrosion even in a high temperature steam environment, have been developed over the world. Several concepts, such as application of a coating to a conventional zirconium cladding or a multi-metal layer cladding, have been studied. In a short term, it is necessary to study coating technology, but it also has disadvantages such as peeling of the coating layer at accident and difficulty of formation of uniform coating layer at 4-m long cladding. From the long-term point of view, it is necessary to study FeCrAl alloys that can replace existing zirconium cladding.

FeCrAl alloys are known to have excellent oxidation resistance in a high temperature steam environment. They also have resistance to corrosion during primary water environment and to hydrogen embrittlement. But the optimized composition for light water reactor environments has not been determined yet.

In this study, first, the composition of FeCrAl alloy optimized for PWR environment will be determined and then we will evaluate the high temperature oxidation resistance in steam environment, long-term corrosion resistance in primary water, irradiation resistance and mechanical properties of the final FeCrAl alloy with minor elements. The development plan is divided into three stages as shown in Fig. 1. In this paper, the manufacturing process of FeCrAl plate sample and the results of high-temperature steam corrosion test are described and the future plan is briefly introduced.

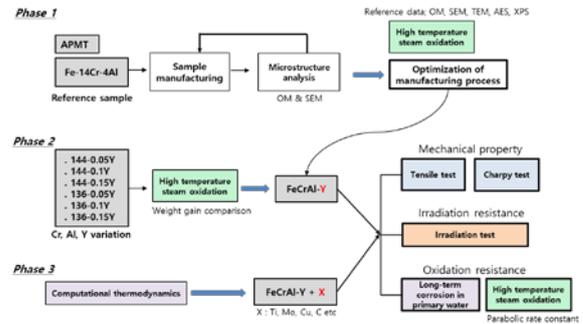


Fig. 1. Schematic diagram of research plan

2. Sample manufacturing process

FeCrAl alloy with 14 wt. % Cr and 4 wt. % Al composition was manufactured as a reference sample for the optimization of the manufacturing process. Cr has maximum content limitation of 20 wt. % in order to prevent the formation of α' -Cr phase and Laves-phase. And at least 13 wt. % Cr is necessary for stable Cr oxide scale formation in the primary water. For Al, there is a maximum content limitation of 6 wt. % to ensure fabricability and at least 4 wt. % Al is necessary for the uniform Al_2O_3 formation [2,3,4]. As a starting point, 14 wt. % Cr and 4 wt. % Al was selected in those composition ranges.

An ingot with a width of 150 mm, a length of 320 mm and a thickness of 80 mm was cast by vacuum induction melting. Since the main alloy elements in the ingot may not be uniformly distributed in the matrix, the ingot was homogenized at 1150 °C for 2 hours. Hot rolling and annealing were performed after the homogenization. It is important that the hot rolling and annealing process are performed at appropriate temperature and time, because they greatly affect the grain size and shape.

Previous study suggested that the grain size of the specimen annealed at 700 °C for one hour after hot rolling at 700 °C is minimized to less than 100 μm and other researcher suggested that the elongated grain is removed when the total thickness reduction is 93%. Therefore, in this study, homogenized ingot was annealed at 700 °C for 1 hour after hot rolled with a 92.5% total thickness reduction [5].

The schematic diagram of the sample manufacturing process is shown in Fig. 2.

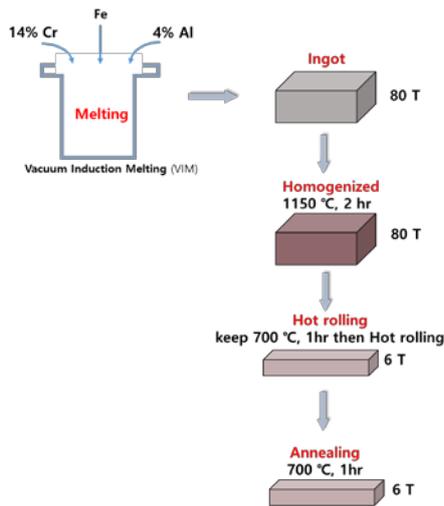


Fig. 2. Schematic diagram of FeCrAl plate sample manufacturing process.

3. Experimental methods

The grain size and shape of two cross-sections perpendicular to the rolled surface and the rolled surface of the Fe14Cr4Al alloy were analyzed using optical microscopy (Nikon LV150N). A scanning electron microscope was used to analyze whether the particles or precipitates existed in the grain boundary or matrix.

The specimens were cut by a low speed saw at the central part of the mother sample and polished. Each of the polished specimens was etched with a solution containing 45 ml of glycerol, 30 ml of hydrochloric acid and 15 ml of nitric acid, and then ultrasonic cleaning with ethanol and distilled water was conducted. The diameter and size of grains were measured by the intercept method with reference to ASTM E112-13.

To evaluate the initial oxidation behavior in a high temperature steam environment, corrosion testing was carried out for 200 ~ 400 seconds by injecting a specimen into a furnace with 1200 °C steam environments.

4. Results & discussion

4.1. As-received sample analysis

Figure 3 shows the optical microscopy images of the rolled surface, cross section 1 and cross section 2. In the rolled surface, recrystallized grains are well formed. But it can be seen that elongated grains exist in the middle part of the both cross-sections. These elongated grains depend on the number of rolling pass [6] and depend on the Al content [7].

The elimination of such elongated grains is one of the main goals. Further optimization of the manufacturing process is being carried out.

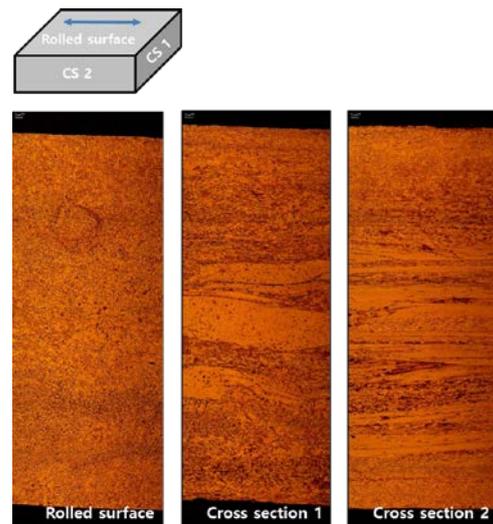


Fig. 3. Optical microscope images (x50 magnification) of rolled surface and both two cross sections that are each perpendicular to the rolled surface.

4.2. High temperature steam oxidation test

High temperature steam oxidation testing was performed for Fe14Cr4Al and commercial APMT (21Cr-5Al-3Mo). The polished Fe14Cr4Al and APMT samples were exposed at 1200 °C for 371, 250 seconds, respectively. Figure 4 shows the surface images of the both oxidized samples. The surface morphology of Fe14Cr4Al sample divided into red and blue regions. Further analysis is underway for these regions.

The weight gain results of Fe14Cr4Al and APMT are shown in the Fig. 5. Both alloy showed higher oxidation resistance than Zr-Nb-Sn alloy at 1200 °C steam environment. These are basic data to be compared with future FeCrAl-Y and FeCrAlY-X. Additional experiments will be carried out with increasing the exposure time.

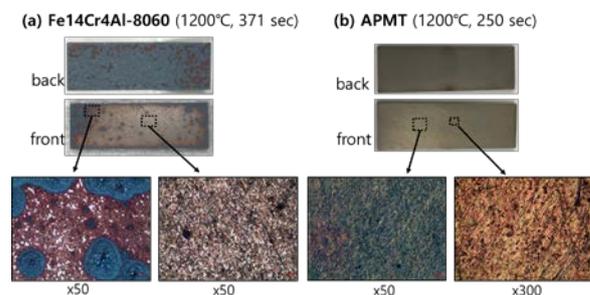


Fig. 4. Surface images of Fe14Cr4Al and APMT exposed in a 1200 °C steam environment.

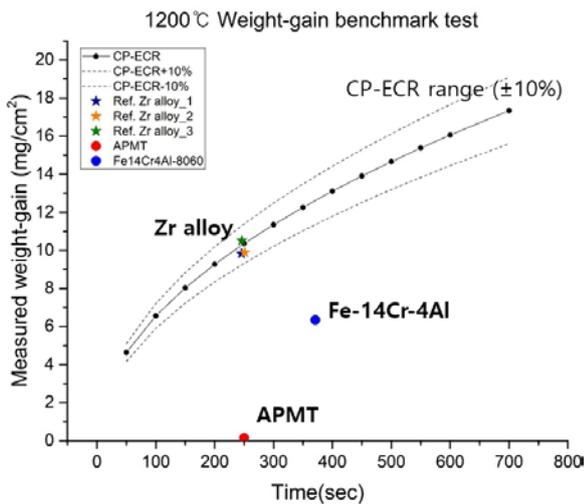


Fig. 5. Weight gain bench mark test of FeCrAl, APMT and Zr alloy corroded at 1200 °C steam environment.

4.3. Future work

Additional microstructure analysis and high-temperature steam oxidation test will be conducted on the reference sample, Fe14Cr4Al. As shown in Fig. 1, in stage 2, the high temperature oxidation testing will be performed on 14Cr4Al and 13Cr6Al alloy doped with yttrium ranging from 0.05 wt. % to 0.15 wt. %, and the parabolic rate constants, k_p for each alloy composition will be compared. The composition with the highest steam oxidation resistance will be selected as the stage 3 sample. In stage 3, FeCrAl-Y alloy doped with an additional minor element will be fabricated to evaluate mechanical properties and irradiation resistance.

5. Summary & conclusions

This study consists of three stages and stage 1 was being studied. The Fe14Cr4Al plate alloy was manufactured, and compared with APMT and Zr-Nb-Sn alloys by exposure at 1200 °C in steam environment. The summary is as follows.

1. To fabricate the FeCrAl plate alloy, the ingot was casted by vacuum induction melting, homogenized at 1150 °C for 2 hours and hot-rolled with a total thickness reduction rate of 92.5%, and annealed at 700 °C for 1 hour.
2. Elongated grains were observed in the middle of two cross-sections perpendicular to the rolled surface. Further optimization of the manufacturing process is being carried out.
3. Polished Fe14Cr4Al and APMT specimens were exposed at 1200 °C in steam environment for 371 and 250 seconds, respectively. It is confirmed that the

both Fe14Cr4Al and APMT alloy have better oxidation resistance than Zr-Nb-Sn alloys.

4. Unlike APMT, the surface of Fe14Cr4Al is divided into red and blue regions. This means that the surface is oxidized locally at the initial stage of oxidation kinetic. Further analysis is underway for these regions. Additional experiments will be carried out with increasing the exposure time.
5. In stage 2, the high temperature oxidation testing will be performed on 14Cr4Al and 13Cr6Al alloy doped with yttrium ranging from 0.05 wt. % to 0.15 wt. %. The parabolic rate constants for each alloy will be compared and the composition with highest steam oxidation resistance will be selected as the stage 3 sample.

6. In stage 3, FeCrAl-Y alloy doped with an additional minor element will be evaluated mechanical properties and irradiation resistance.

ACKNOWLEDGMENT

This work was supported by the National Research Foundation of Korea(NRF) grant funded by the Korean government(MSIP:Ministry of Science, ICT and Future Planning) (No. 2017M2A8A4056239), “Human Resources Program in Energy Technology” of the Korea Institute of Energy Technology Evaluation and Planning (KETEP), granted financial resource from the Ministry of Trade, Industry & Energy, Republic of Korea. (No. 20164010201000).

REFERENCES

- [1] P. Hofmann, Current knowledge on core degradation phenomena, a review. Journal of Nuclear Materials Vol.270, p.194-211, 1999.
- [2] S. Kobayashi, T. Takasugi, Mapping of 475°C embrittlement in ferritic Fe-Cr-Al alloys. Scripta Materialia Vol.63, p.1104-1107, 2010.
- [3] J. Bischoff et al., "Proceedings of the Third Meeting of the OECD-NEA Expert Group on Accident Tolerant Fuels for LWRs, 3-5 March 2015, OECD-NEA HQ", Organisation for Economic Co-Operation and Development, 2015
- [4] Y. Yamamoto, "Development and quality assessments of commercial heat production of ATF FeCrAl tubes", Oak Ridge National Lab. (ORNL), Oak Ridge, TN, United States, 2015.
- [5] Y. Yamamoto et al., Development and property evaluation of nuclear grade wrought FeCrAl fuel cladding for light water reactors. Journal of Nuclear Materials Vol.467, p.703-716, 2015.
- [6] Z. Sun, Y. Yamamoto, Processability evaluation of a Mo-containing FeCrAl alloy for seamless thin-wall tube fabrication. Materials Science and Engineering: A 700, p.554-561, 2017.

[7] J. Weaver, E. Aydogan, N. A. Mara, S. A. Maloy,
"Nanoindentation of Electropolished FeCrAl Alloy Welds",
United States, 2017.