Evaluation of Fe-20Cr-2Si Oxidation Resistance in 1200 °C Steam Environment

Joonho Moon^a, Sungyu Kim^a, Ji Hyun Kim^b, Michael P. Short^c, Chi Bum Bahn^{a*}

^aSchool of Mechanical Engineering, Pusan National University, 2, Busandaehak-ro 63beon-gil, Geumjeong-gu,

Busan, 46241

^bDepartment 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,

^cDept. 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

The zirconium alloy fuel cladding has being used in nuclear power plants. Under the nuclear power plant operating condition, the zirconium alloy fuel cladding has been showing very good performance. However, under a severe accident condition, similar to the Fukushima accident, the zirconium alloy fuel cladding can release a large quantity of hydrogen gas due to rapid oxidation, and cause hydrogen explosion. In order to prevent this, accident tolerant fuel (ATF) cladding has been studied in many countries [1-2]. Multi-metallic layered composite (MMLC) cladding is a new ATF cladding design. MMLC is consisted of zirconium alloy, FeCrSi alloy and buffer materials in between them. Among these materials, the FeCrSi alloy is located at the outermost part of MMLC and acts as an oxidation barrier. In our previous study, the Fe20Cr2Si alloy has been proven to maintain excellent resistance in steam-Ar environment until 100 sec [3]. However, longer-term testing than 3 hr is needed to confirm that MMLC cladding can be considered as ATF cladding. Therefore, we conducted high temperature oxidation tests in steam-Ar environment until 24 hr.

2. Experimental Methods

Before the high temperature oxidation test, the specimens of Fe20Cr2Si alloy were prepared as 30×20 \times 2 mm coupons. These specimens polished using SiC paper up to P2400 and cleaned with ethanol, acetone, and pure water in series. The oxidation tests were conducted referring to NRC Reg. guide DG-1262 [4] in the resistance heating tube furnace. In order to increase the specimen heating speed, once a furnace was heated up to 1200 °C and stabilized, the specimen was inserted into the furnace. It was oxidized at 1200 °C for about 100 sec, 10 min, 1 hr, 3 hr, 8 hr, and 24 hr. After the oxidation test, the specimen was taken out of the furnace and cooled down under air environment. During the oxidation tests, steam and Ar gas were supplied into the furnace at 500 cc/hr, respectively. The mixture gas was heated up to 300 °C before injecting into the furnace in order to prevent the water condensation.

3. Results and Discussion

After the oxidation tests, the oxide layers were measured and observed using Scanning Electron Microscope (SEM) with Dispersive X-ray Spectroscopy (EDS). The weight change result is shown in Table I.

Table I: Weight change of Fe20Cr2Si alloy over time

	100 sec	10 min	1 hr	3 hr	8 hr	24 hr
1	0.17	0.27	0	-0.76	-0.94	1.38
2	0.19	0.39	0.23	-0.48	0.39	1.70
3	0.27	-	-	-	-	1.39
4	0.17	-	-	-	-	-
Spall ing	Х		О			

In Table I, it is observed that Fe20Cr2Si alloy has a slight weight gain less than 1hr test. However, in more than 1hr test, weight loss is occurred. It is considered due to spalling out of some oxide layers during cool down. In order to observe the surface of specimens, it was conducted the SEM-EDS analysis of surface. Figure 1 shows the surface shape of specimens and Fig. 2 shows the surface composition of specimens.



Fig. 1. Surface shape of Fe20Cr2Si alloy over time after oxidation test (Magnification: X50)

As shown in Fig 1, the area fraction of oxide spalling increases with time until 8 hr. However, in the 24 hr test specimen, more oxide layers still remain compared with the 8 hr test specimen. Figure 2 shows Cr oxide, Si oxide, and base metal. The base metal area is the oxide spalling area. It appears that there is no change in the surface oxide composition over time.



Fig. 2. EDS mapping results on the surface of Fe20Cr2Si alloy over time after oxidation test

In order to determine the oxide thickness and composition, the SEM-EDS analysis of the cross-section was conducted. The oxide thickness and shape are shown in Fig. 3.





Fig. 3. Comparison of oxide thickness of Fe20Cr2Si alloy over time

Figure 3 shows that oxide layers still exist until the 24 hr test. The oxide thickness increases up to about 20 μ m for the 24 hr test. The oxide thickness of Fe20Cr2Si 24 hr test is about half of the oxide thickness of Ref. Zr-Nb-Sn alloy 100 sec test. Although the oxide thickness increases with time, the oxidation resistance of Fe20Cr2Si alloy is superior to Ref. Zr-Nb-Sn alloy.

Figure 4 shows the composition of oxide layers. It shows the Cr oxide and Si oxide in all specimens. It is thought that the Cr oxide and Si oxide of Fe20Cr2Si alloy are not either evaporated or dissolved significantly until 24 hr. The previous study reported that Cr oxide growth rate is faster than Cr oxide evaporation rate in N-O₂-H₂O environment [5], which appears to be similar to our case. Therefore, it is thought that Cr oxide continuously grows due to lower Cr oxide evaporation rate and the Si oxide is prevented from steam contact due to the thick Cr oxide. Since the Cr oxide and Si oxide remain until 24 hr, it is considered that Fe20Cr2Si alloy has enough steam oxidation resistance to be considered as accident-tolerant alloy.



Fig. 4. EDS mapping results on the cross-sectional images of oxide layers of Fe20Cr2Si alloy over time

4. Conclusions and Future Work

In this study, the oxidation resistance of Fe20Cr2Si alloy is evaluated in 1200 °C steam-Ar environment until 24 hr. In less than 1 hr tests, the Fe20Cr2Si alloy has excellent oxidation resistance. In 1 hr to 24hr tests, although partial oxide spalling occurs during cool down, it has superior oxidation resistance to Ref. Zr-Nb-Sn alloy. It is also observed that Cr oxide and Si oxide layers, acting as an oxidation barrier, remain until 24 hr. Therefore, it is concluded that Cr oxide continuously grows due to lower Cr oxide evaporation rate and the Si oxide is prevented from steam contact due to the thick Cr oxide.

In order to know the oxide growth mechanism, it is planned to conduct the analysis of microstructure of Cr oxide and Si oxide.

ACKNOWLEDGEMENT

This work was financially supported by the International Collaborative Energy Technology R&D Program (No. 20168540000030) of the Korea Institute of Energy Technology Evaluation and Planning (KETEP) which is funded by the Ministry of Trade Industry and Energy and was supported by "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. 20184010201660).

REFERENCES

[1] Kim, Hyun-Gil, Yang, J. H., Kim, W. J., and Koo, Y. H., 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] Moon, Joonho, Kim, Sungyu, Park, Won Dong, Kim, Tae Yong, McAlpine, Samuel Westcott, Short, Michael P., Kim, Ji Hyun, Bahn, Chi Bum, Initial oxidation behavior of Fe-Cr-Si alloys in 1200° C steam, Journal of Nuclear Materials, Vol.513, p.297, 2019.

[4] U.S. Nuclear Regulatory Commission (NRC), DRAFT REGULATORY GUIDE DG-1262, TESTING FOR POSTQUENCH DUCTILITY

[5] Yamauchi, Akira, Kazuya Kurokawa, and Hideaki Takahashi., Evaporation of Cr₂O₃ in atmospheres containing H₂O, Oxidation of metals Vol.59, p.517, 2003