Si Effect on High Temperature Steam Oxidation of FeCrSi Alloy

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

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. It 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 responsible for oxidation protection. In our previous study, the Fe20Cr2Si alloy has been proven to maintain excellent resistance in steam-Ar environment until 24 hr. Furthermore, the high temperature oxidation resistance of FeCrSi alloys shows different characteristics depending on the Cr content [3-4]. Previous study showed that the addition of Si increases the oxidation resistance [5]. However, it is not verified in high temperature steam environment such as loss-ofcoolant accident conditions. Therefore, we conducted the oxidation tests in high temperature steam-Ar environment until 24 hr in order to check the Si effect in FeCrSi alloy.

2. Experimental Methods

The FeCrSi alloy specimens were prepared as 30 \times 20×2 mm coupons. These specimens were polished using SiC paper up to P2400 and cleaned with acetone, ethanol, and pure water in series. The oxidation tests were conducted at 1200 °C in the resistance heating tube furnace, referring to NRC Reg. guide DG-1262 [6]. 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 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, after heated to 300 °C. The chemical composition of FeCrSi specimens is shown in Table I. The Fe20Cr and Fe20Cr1Si specimens were analyzed using Laser Induced Breakdown Spectroscopy (LIBS), the

Fe20Cr2Si specimen was analyzed using Direct Current Plasma-Atomic Emission Spectroscopy (DCP-AES).

|--|

	Fe	Cr	Si	Method
Fe20Cr	Bal.	20.42	0.00	LIBS
Fe20Cr1Si	Bal.	20.39	1.14	LIBS
Fe20Cr2Si	Bal.	20.06	1.95	DCP-AES

3. Results and Discussion

After the oxidation test, in order to check the specimen condition, photos of the specimens were taken. Figures 1 and 2 show the shape of specimens after the oxidation test.



Fig. 1. Surface morphology of FeCrSi alloys after oxidation test and cooling for 1 hr



Nodular oxidation

Fig. 2. Comparison of surface morphology of FeCrSi alloys with increasing the Si content after 24 hr test

In Figs. 1 and 2, the nodular oxidation area is observed in Fe20Cr and Fe20Cr1Si. In the Fe20Cr specimen, it is observed that the total area of the nodular oxidation increases over time. In the Fe20Cr1Si specimen, the nodule shape is observed only near the edge in 24 hr test. The nodule shape is not observed in Fe20Cr2Si. This result suggests that the addition of Si in FeCr alloy is effective to prevent the nodular oxidation occurrence.

In order to check the oxide composition of specimens, The SEM-EDS analysis of cross-section was conducted. The EDS mapping result of Fe20Cr specimen is shown in Figs. 3 and 4.



Fig. 3. EDS mapping results on the cross-sectional images of oxide layers of Fe20Cr alloy over time



Fig. 4. EDS mapping results on the cross-sectional images of nodular oxidation area of Fe20Cr in 8 hr test

Figure 3 shows the composition of non-nodule oxide layer of Fe20Cr alloy. Only Cr oxide is observed until 24 hr. However, as shown in Fig. 4, Fe oxide and Cr oxide are observed in the nodule oxide layer. The top of the nodule oxide area consists of Fe oxide; the bottom area consists of Fe oxide and Cr oxide mixture. Also, in the edge area of nodule oxide, it is observed that outside oxide area consists of Fe oxide and inside oxide area consists of Cr oxide.

Figure 5 shows the EDS mapping result of Fe20Cr1Si specimen.



Fig. 5. EDS mapping results on the cross-sectional images of oxide layers of Fe20Cr1Si alloy over time

As shown in Fig. 5, Two distinct oxide layers are observed: outer Cr oxide and inner Si oxide layer. The Si oxide layer thickness increases over time, which is the same trend as observed in the Fe20Cr2Si results.

Based on SEM-EDS analysis results of Fe20Cr and Fe20Cr1Si, it is thought that the addition of 1 wt% Si effectively prevents the formation of the Fe oxide and increases the oxidation resistance of FeCr alloy.

4. Conclusions and Future Work

This study evaluates the Si effect on the high temperature steam oxidation of FeCrSi alloy. In the Fe20Cr alloy, the local nodular oxidation occurs, and the magnitude of that increases over time. In the Fe20Cr1Si alloy, it is observed that the small nodular oxidation in the edge area only after the 24 hr test. This nodular oxidation was not observed in Fe20Cr2Si until 24hr [4]. Based on this result, it is thought that the addition of Si in FeCr alloy is effective to increase the oxidation resistance in high temperature steam-Ar environment. The reason for the increase in the oxidation resistance is that the addition of Si prevents the formation of the outer Fe oxide by forming the continuous Si oxide layer at the interface between metal matrix and oxide. In order to know detailed oxidation resistance mechanism of the Si element addition, it is needed to conduct additional analysis, such as XRD, TEM etc.

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] Moon, Joonho, Kim, Sungyu, Short, Michael P., Kim, Ji Hyun, Bahn, Chi Bum, Evaluation of Fe-20Cr-2Si Oxidation Resistance in 1200 °C Steam Environment, Proceedings of Transactions of the Korean Nuclear Society Spring Meeting, May 23-24, 2019, Jeju, Korea,

[5] Robertson, J., Manning, M. I., Healing layer formation in Fe–Cr–Si ferritic steels. Materials Science and Technology, Vol.5, p.741, 1989

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