Study on the Reducing Separation of Ceramic Coating Layer Used for Melting Crucible of Metal Fuel

Ki-Won Hong[a,b], Sun-Ig Hong[a], Ki-Hwan Kim[b]

[a] *Materials Science & Engineering, Chungnam National University, Daejeon, 34134, Republic of Korea*
[b] *SFR Nuclear Fuel Development Division, Korea Atomic Energy Research Institute, Daejeon, 34507, Republic of Korea*
Corresponding author: khkim2@kaeri.re.kr (Ki-Hwan Kim)

1. Introduction

Research on the prevention of interaction has progressed since 1990s because graphite crucible interacts with uranium alloy at high temperature, which causes a contamination of the fuel slugs [1-4]. Reaction testing of ceramic plasma-spray coated with U-10wt.%Zr-5wt.%RE has progressed to the evaluation of the protective performance of ceramic materials. Y_2O_3 was selected as protective ceramic material and sprayed onto graphite substrates [5-8]. However, separation problem was found due to its difference of coefficient of thermal expansion (CTE).In this study, Niobium which has similar CTE value with Y_2O_3 was applied as a new substrate and SiC layer was also applied as a bond coating layer. Interaction research progressed through a melt reaction test at 1470°C. SEM/EDS investigations were conducted to evaluate the micro-structural features of the coatings. Yttrium oxide on niobium crucible shows a better separation resistance for U-10wt.%Zr-5wt.%RE melt.

2. Methods and Results

2.1 Thermal Cycling Test

150 microns of a yttrium oxide layer were formed on the niobium substrate and the SiC-layer. Also, SiC-layer was formed on graphite substrate by CVD methods. Y_2O_3 coating layers were applied using a plasma-spray coating method. Thermal cycling test was carried out in 5 cycles. The applied temperature cycle is shown in Fig. 1.

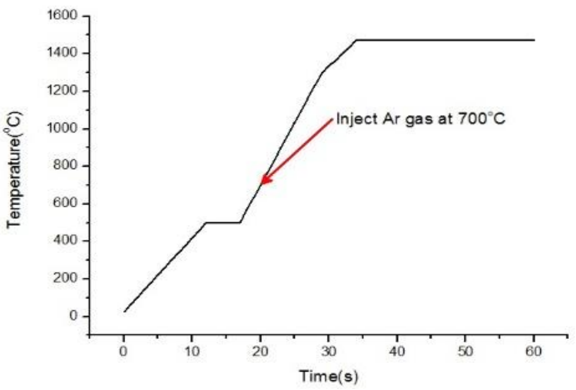

Fig. 1. Applied temperature cycle used for thermal cycling test.

Fig 2. shows as-coated ceramic coating layer and after annealed ceramic coating layer. In Fig. 2-(a) and (b), Y_2O_3 coating layer were perfectly formed on niobium substrates and SiC layer. Some pores are observed in coating layer. Photographs of Y_2O_3 coating layers on niobium substrates after a thermal cycling test are shown in Fig. 2-(c), (e), and (g). Any cracks and separations weren't observed in all niobium specimens. Relatively, Y_2O_3 coating layers on SiC layers after a thermal cycling test didn't show a good performance. Some cracks were observed in coating layer but separation weren't observed in fig 2-(d), (f), and (h).

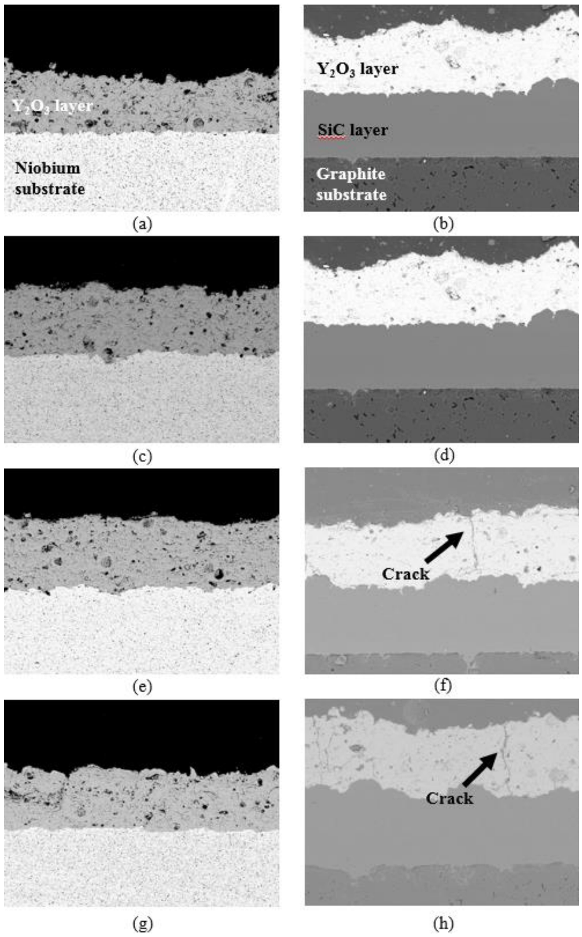

Fig. 2. Cross-sectional BSE micrographs showing the plasma-sprayed coating layer on niobium substrate and SiC layer; (a) As coated (150µm, on niobium substrate), (b) As coated (150µm, on the SiC layer), (c),(d) after 1 cycle, (e), (f) after 3 cycles, and (g), (h) after 5 cycles.

2.2 Melt Reaction test

Photographs of the niobium crucible after a melt reaction test and cut niobium crucible are shown in Fig. 3-(a) and (b). In Fig 3-(b), a slight gap between crucible and melts was observed. It is thought to be due to difference of cooling rate between niobium crucible and melts. Also, in Fig 3-(b), reaction area was observed at the top of the melts. It is thought to be due to difference of density between uranium and rare earth elements which has very strong reactivity. Finally, sections to be SEM-EDS analyzed are shown in fig 3-(b).

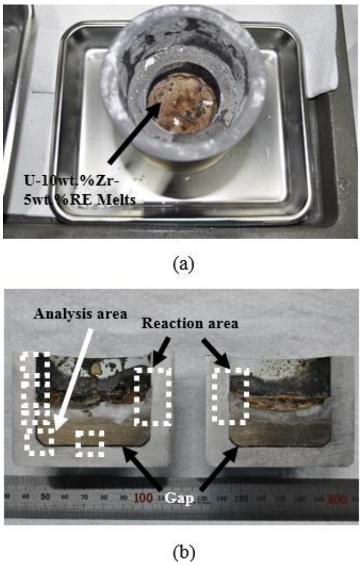

Fig. 3. Image of crucible after melt reaction test and cut niobium crucible.

SEM micrographs of the niobium crucible after a melt reaction test are shown in Fig. 4. In Fig. 4-(a), the top section, slight cracks were observed but there was no separation on the niobium crucible and condition of coating layer was generally good. In Fig. 4-(b), the boundary section, there was no coating layer and niobium was reacted by uranium and rare earth elements. Thin reaction later was observed between niobium and U-10wt.%Zr-5wt.%RE melts. Finally, lower section of boundary, curved section, and bottom section are shown in Fig. 4-(c), (d), and (e). In Fig. 4-(c), just one crack and slight separation between niobium crucible and coating layer were observed. Plasma spray coated Y_2O_3 coating layer shows good performance but separated by penetration of rare elements through destruction of coating layer in the boundary section. Also, coating layers of curved section and bottom section were shows good performance.

EDS mapping of the boundary section and lower section of boundary are shown in Fig. 5 and Fig. 6. In Fig. 5, niobium crucible was reacted only uranium elements. Similar results of EDS analysis were observed in Fig. 5-(c) and (d). On the other hand, the rest of the area was filled with rare earth elements. In Fig. 6, any trace of infiltration wasn't observed inside of Y_2O_3 coating layer. As mentioned in fig. 4, trace of infiltration due to rare earth elements was observed between niobium crucible and coating layer. It is thought that the rare earth elements infiltrated due to the relatively weak bonding force due to the destruction of the coating layer at the boundary section.

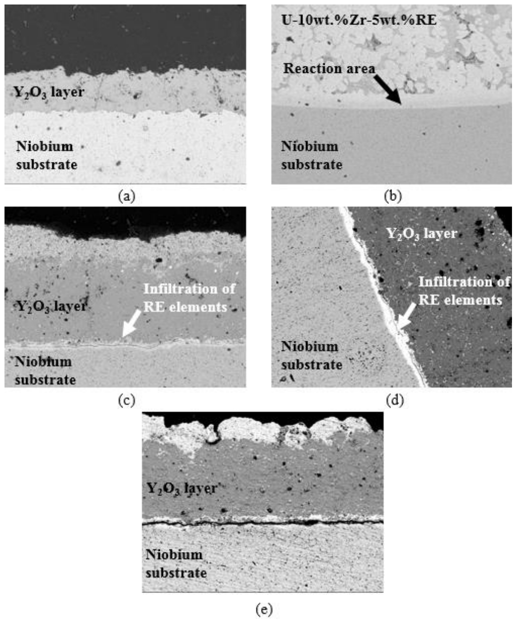

Fig. 4. Cross-sectional BSE micrographs showing the surface of Y_2O_3 coating layer after melting test at 1470ºC; (a) the top section, (b) the boundary section, (c) the lower section of boundary, (d) the curved section, and (e) the bottom section.

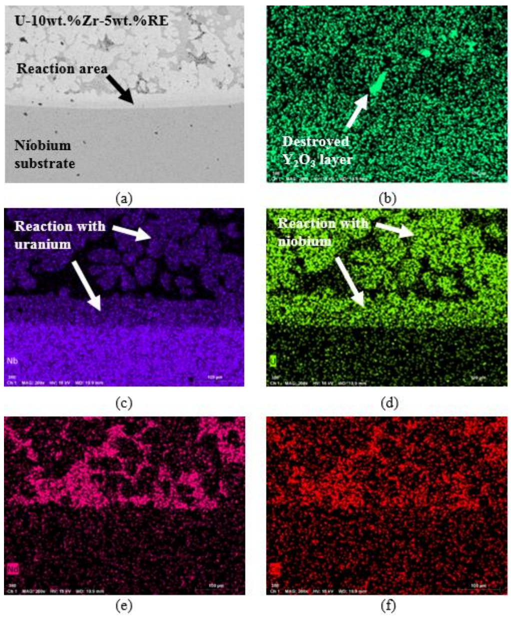

Fig. 5. EDS-mapping test of the cross-section of the boundary section; (a) original image of the boundary section, (b) yttrium element, (c) niobium element, (d) uranium element, (e) neodymium element, and (f) cerium element.

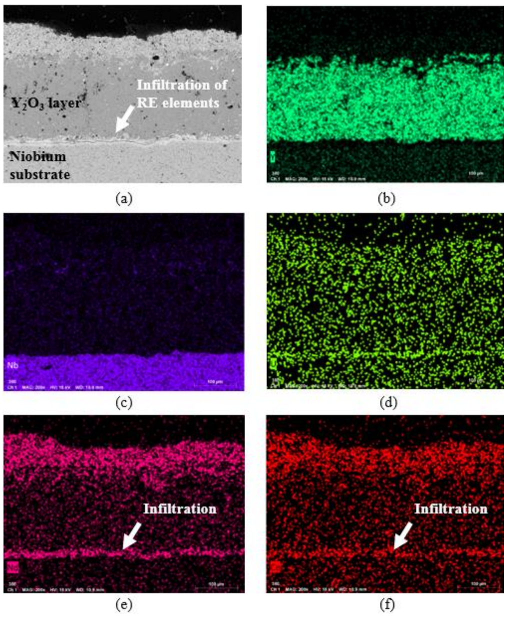

Fig. 6. EDS-mapping results of the cross-section of the lower section of boundary; (a) original image of the lower section of boundary, (b) yttrium element, (c) niobium element, (d) uranium element, (e) neodymium element, and (f) cerium element.

3. Conclusions

The Y_2O_3 coating layer using plasma-spray coating method layer on a niobium substrate and SiC layer shows a good adhesion performance in thermal cycling test. However, some cracks were detected in the Y_2O_3 coating layer on the SiC layer. It is thought to the difference of CTE value between Y_2O_3 coating layer and the SiC layer. In the melt reaction test, Y_2O_3 coating layer shows good resistance with rare earth elements. However, trace of infiltration due to rare earth elements was observed between niobium crucible and coating layer. It was caused that the infiltration of rare earth elements. If the separation by difference of density and immiscibility of rare earth elements are prevented in the casting process, it is thought that the failure of the coating layer at boundary section can be prevented.

REFERENCES

[1] Robert B. Heimann, Plasma Spray Coating, Principle and Applications, Wiley-VCH, 1996.

[2] L. Pawlowski, The Science and Engineering of Thermal Sparay Coatings, John Wiley & Sons Ltd, Chichester, England, 1995.

[3] X.Q. Cao, R. Vassen, D. Stoever, J. European Ceramic Society, 24 (2004) 1-10.

[4] E. Celika, I. Ozdemir, E. Avci, Y. Tsunekawa, Surface & Coatings Tech. 193 (2005) 297– 302.

[5] Ki Hwan Kim, Chong Tak Lee, Chan Bock Lee, R.S. Fielding, J.R. Kennedy, Journal of Nuclear Materials, 2013.

[6] Sung Ho Lee, Choon Ho Cho, Yoon Sang Lee, Han Soo Lee, Jeong-Guk Kim, Korean, J. Chem. Eng., 27(6) (2010) 1786-1790.

[7] Ki Hwan Kim, Chong Tak Lee, Chan Bock Lee, R.S. Fielding, J.R. Kennedy, Thin Solid Films, 519 (2011) 6969–6973.

[8] J.W. Koger, C.E. Holcombe, J.G. Banker, Thin Solid Films, 39 (1976) 297-303.