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A Eutectic Melting Study of Double Wall Cladding Tubes of FeCrAl and Zircaloy-4

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1. Introduction

The development of oxidation resistant fuel cladding has been one of the major research topics of accident tolerant fuel development[1,2]. The FeCrAl alloys have been considered one of the promising candidate materials for the oxidation resistant cladding because several experimental results have demonstrated the excellent steam oxidation resistance of FeCrAl alloys at very high temperatures up to 1200°C[3,4]. Meanwhile, the irradiation embrittlement and inferior neutron economy have been serious drawbacks of FeCrAl alloys[5,6]. Reducing the Cr content below 14-16wt.% or minimizing the cladding thickness as thin as possible are possible solutions to the problems of FeCrAl alloys for accident resistant cladding.

Recently, coating of FeCrAl layers on the Zr alloy cladding tube is being investigated for the development of accident tolerant fuel by exploiting of both the oxidation resistance of FeCrAl alloys and the neutronic advantages of Zr alloys[7-10]. Coating of FeCrAl alloys on Zr alloy cladding tubes can be performed by various techniques including thermal spray, laser cladding, and co-extrusion. Son et al. also reported the fabrication of FeCrAl/Zr ally double wall cladding by the shrink fit method[11]. For the double layered cladding tubes, the thermal expansion mismatch between the dissimilar materials, severe deformation or mechanical failure due to the evolution of thermal stresses can occur when there is a thermal cycling.

In addition to the thermal stress problems, chemical compatibilities between the two different alloys should be investigated in order to check the stability and thermal margin of the double wall cladding at a high temperature. Generally, it is considered that Zr alloy cladding will maintain its mechanical integrity up to 1204°C (2200°F) to satisfy the acceptance criteria for emergency core cooling systems (ECCS). Therefore, maintaining the chemical stability should be confirmed for double walled or coated cladding systems. However, many conceptual designs for double walled or coated cladding have not taken into account of the chemical compatibilities seriously.

In this study, the chemical compatibility between FeCrAl and Zr alloys are investigated by using double wall tubes of Kanthal (FeCrAl) and Zircaloy-4. The formation of eutectic melting was measured by vacuum annealing at various temperatures between 900°C to 1300°C.

2. Experimental Procedures

Kanthal (APMT) with a composition of 69wt.%Fe-21.6wt%Cr-4.9wt%Al and others was used for FeCrAl tubes. Double wall cladding tubes of FeCrAl and Zircaloy-4 were fabricated by the shrink fit method. A Zircaloy-4 tube was inserted into the heated FeCrAl tube and the gap between the two tubes was closed as the temperature of the FeCrAl tube decreased by cooling. The gap spacing was designed to maintain appropriate mechanical bonding by the compressive stress at the operating temperature of the cladding. The double wall cladding tubes were annealed at temperatures ranging from 900°C to 1300°C for 1 hour under vacuum. The microstructures of annealed double wall cladding tubes were observed by scanning electron microscopy and the crystallographic information of reaction phases was obtained by X-ray diffraction analyses. The possible chemical reactions and related eutectic melting were estimated by using a phase diagram between Fe-Zr as shown in Fig. 1[12].

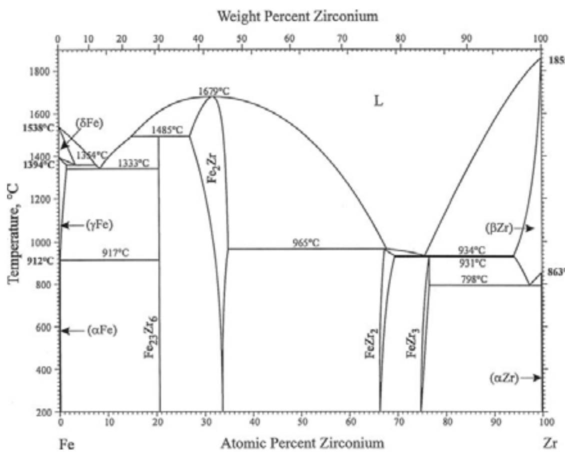


Fig. 1. The Fe-Zr binary phase diagram [12]

3. Results

After annealing for 1 hour at various temperatures from 900°C to 1300°C, the microstructures of double wall cladding tube samples were observed. The appearances of annealed samples could be differentiated depending on melting of the inner layer, Zircaloy-4. From 900°C to 1125°C, melting of the inner layer tube did not occur after annealing for 1 hour under vaccuum. Melting of the inner layer was observed after annealing at tempeatures equal to or higher than 1150°C as presented in Fig. 2.

As a result of interdiffusion of FeCrAl and Zircaloy-4, intermediate reaction phases containing Fe, Cr, Zr formed at the interface of FeCrAl and Zircaloy-4. The phase can be Laves $(Fe,Cr)_2Zr$ because each concentration of Fe, Cr, and Zr is similar to one third in the reaction phase according to energy dispersive spectroscopy (EDS) analyses. As the concentration of Zr in the Fe-Zr system increases over 33 at.%, the melting temperature of the intermediate reaction phase decreases significantly from 1679°C to 965°C. Therefore, eutectic melting can occur even at below 1200°C when enough interdiffusion of Fe and Zr is allowed.

Fig. 2. The melting of Zircaloy-4 inner layer of a double wall cladding tube after annelaing at 1150°C.

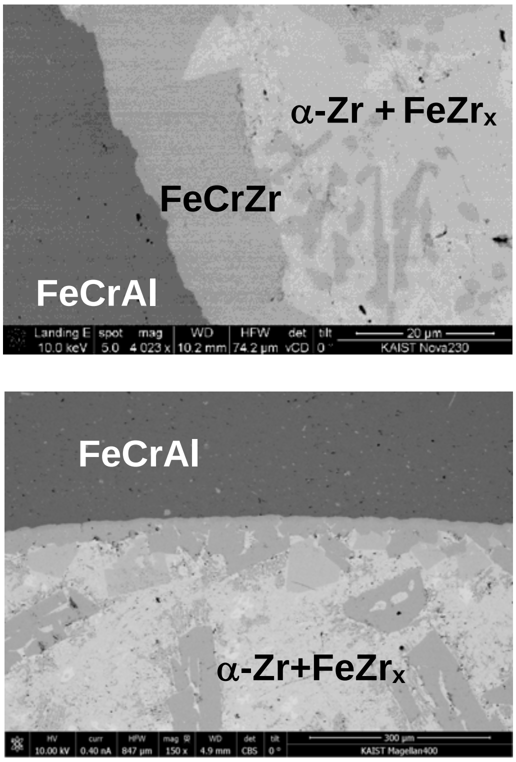
(a) (b)


Fig. 3. SEM cross section microstructures of double wall cladding tubes annealed at (a) 1200°C for 1 hour and (b) 1300°C for 1 hour.

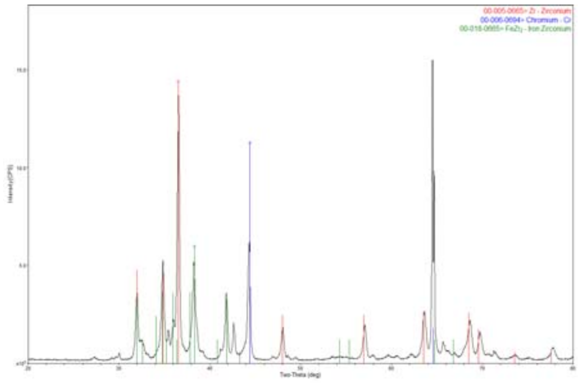


Fig. 4. X-ray diffraction pattern of a double wall cladding tube annealed at 1150°C for 1 hour.

Cross section micrographs of the interfacial area and eutectic melting area presented that beyond a Fe-Cr-Zr layer on FeCrAl, Zr-rich area whose composition is similar to $FeZr_2$ or $FeZr_3$ formed. The existence of $FeZr_2$ in eutectic melting area was confirmed by X-ray diffraction analyses as presented in Fig. 4.

In this study, it is found that the eutectic melting of FeCrAl and Zircaloy-4 occurs at a temperature between 1125°C and 1150°C. Further studies using dilatometry or differential scanning calorimetry will specify the onset temperature at which eutectic melting begins. In order to prevent the eutectic melting of the FeCrAl and Zircaloy-4 tube, diffusion barrier layer should be added between the two layers. Chemically stable ceramic layers or refractory metallic layers can be used as a diffusion barrier.

4. Summary

The eutectic melting behavior of FeCrAl/Zircaloy-4 double wall cladding tubes was investigated by annealing at various temperatures ranging from 900°C to 1300°C. It was found that significant eutectic melting occurred after annealing at temperatures equal to or higher than 1150°C. It means that an additional diffusion barrier layer is necessary to limit the eutectic melting between FeCrAl and Zircaloy-4 alloy cladding tubes.

Acknowledgment

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