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Research status on the barrier coating technologies for preventing Fuel Cladding Chemical Interaction (FCCI) in Sodium Fast Reactor (SFR)

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1. Introduction

Nuclear power plant has been recognized as an essential energy source for future humanity owing to its excellent power generation efficiency. Despite the positive effects of nuclear power plants on the environment, the issue of spent nuclear fuel continues to be a major concern. This is why research on new generation nuclear power plants is being conducted. Sodium Fast Reactor (SFR) is one of next-generation nuclear power plants with the great advantage of being able to reuse the spent nuclear fuel as metal fuel through pyro-processing. In addition to the advantages of recycling, studies on metal nuclear fuel have been actively conducted due to the advantages of high thermal conductivity, low coefficient of thermal expansion, excellent proliferation resistance, good compatibility with sodium, and ease to manufacturing. However, the problem that must be solved in these metal fuels is the Fuel Cladding Chemical Interaction (FCCI) phenomenon. FCCI is an inter-diffusion phenomenon caused by the interface of swelled metal fuel and cladding material in the operating condition of the power plant. Brittle intermetallic compounds are generated from the reaction between fission product such as Ce with cladding material (Fe). The intermetallic can react with Fe to form low melting point eutectic compounds; Fe_2Ce. This leads to degrade both of metal fuel and cladding material. From these backgrounds, Korea Atomic Energy Research Institute (KAERI) developed inner cladding tube coating technology for a past decades. This paper summarizes the current status of coating technology to prevent FCCI and introduces the coating technology for the inner surface of the cladding tube carried out by KAERI so far. Through this, research topics to prevent the FCCI phenomena were suggested for the future study.

2. Status of barrier coating technology

2.1 Barrier coating studies reported by other researchers

Past studies related to FCCI barrier coating or deposition have focused on developing an optimal process for forming an excellent barrier coating layer against FCCI. S. H. Jee et al. [1] reported that they formed FCCI barrier by depositing a Zirconium Oxynitride (ZON) thin film on HT9 using Metalorganic Chemical Vapor Deposition (MOCVD). They also confirmed that the thickness and density of the thin film were dependent on the hydrogen flow, and based on this, a sound barrier performance was confirmed in the diffusion couple test with U-10Zr (wt%). In addition, the FCCI resistance was improved when the microstructure of the Zr thin film was changed by using hydrothermal crystallization on the Zr-deposited specimen using RF magnetron sputtering [2]. K. Sridharan et al. [3-5] used Electrophoretic Deposition (EPD) to coat the inner side of HT9 and T91 cladding tubes (ID: 4 mm, Length: 12 in.) with TiO_2, Yttria-Stabilized Zirconia (YSZ), V_2O_3, Ti, Zr, V, and TiN. To determine the direct cause of the barrier coating quality, microstructure analysis was performed by changing the colloidal solution particle size, voltage, current, additives, and sintering conditions. Also, to check the barrier performance of the coated material, they conducted a diffusion couple test with Ce similar to that of nuclear fuel to evaluate which type of coating material has excellent FCCI resistance. F. Khatkhatay et al. [6] formed thin film coating on pure Fe by using Pulsed Laser Deposition (PLD). They reported that TiN and ZrN coatings provide reliable diffusion barrier characteristics against Ce and possibly other lanthanide fission products. W. Lo et al. [7] reported that they selected barrier material as V_2C and Pack Cementation Diffusion Coating (PCDC) methods as coating process. They demonstrate that a V_2C layer with a thickness of less than 5 μm can effectively eliminate the FCCI between cerium and HT9 steel. S. Huang et al. [8] claimed that the vanadium carbide (VC) deposited by MOCVD process has excellent barrier performance on diffusion couple test with pure Ce. And they also demonstrate the smaller grain size of coating layer provide a better performance. D. Kim et al. [9] reported that they deposit the CrN coating on HT9 disk specimen by using RF magnetron sputtering. They also claimed that the ratio of argon and nitrogen gas flow rates is very effective on the quality of barrier. Table 1 indicates the summarize of research status regarding FCCI barrier coating/deposition. Table 1 indicates the summarize of research status regarding FCCI barrier coating/deposition.

Table.1. Research trends of FCCI barrier coating/deposition process

Ref. No.	Year	Method	Material	Sample type	FCCI resistance
[1]	2012	MOCVD	ZON (Zirconium Oxynitride)	HT9 disk	U-10Zr
[2]	2013	RF magnetron sputter + Hydrothermal crystallization	Zr	HT9 disk	Ce-La
[3-5]	2013	EPD	TiO_2, YSZ, V_2O_3, Ti, Zr, V, TiN	HT9 & T91 Inner surface of tube (ID: 4mm, 12 in)	Ce
[6]	2013	PLD	TiN, ZrN	Pure Fe plate	Ce
[7]	2014	PCDC	V_2C	HT9 duct	Ce
[8]	2017	MOCVD	VC, V_2C	HT9 plate	Ce
[9]	2018	RF magnetron sputter	CrN	HT9 disk	Ce-La

2.2 Barrier coating technology of KAERI

Among various barrier coating/deposition techniques that can prevent FCCI, KAERI selected the method of combined Cr electro-plating and plasma assisted nitriding techniques. Because the actual shape of the target application is a long tube with an ID of 7~8 mm and a length of 3~4 m. One of the main practical limitations of many barrier coating methods is that they are cannot be used to effectively deposit coatings on the inner surfaces of long tube shape cladding geometries.

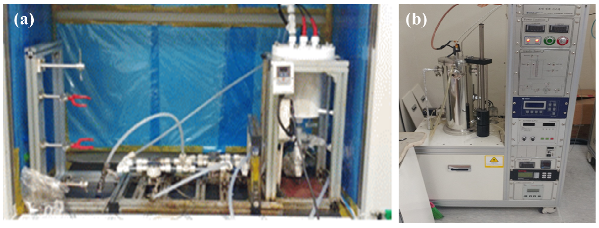

Fig.1. (a) Cr electro-plating equipment and (b) Plasma assisted nitriding equipment

Fig. 1 (a) shows the Cr electro-plating equipment for inside surface of tube, and (b) shows the plasma assisted nitriding set up for this study. A HT9 was used as the substrate material for barrier coating. First, the Cr layer was preferentially plated using Cr electro-plating for the composite layer barrier plating, and then the CrN layer was formed using plasma assisted nitriding.

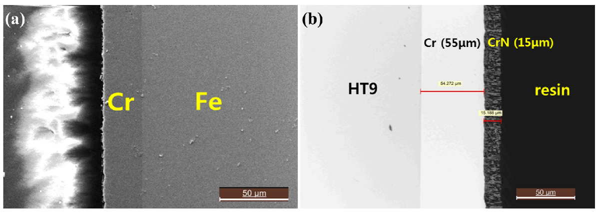

Fig.2. (a) Cr coating and (b) CrN composite coating

Fig. 2 (a) and (b) indicate the cross-sectional microstructure of the Cr electro-plated specimen and the CrN composite coating, respectively. Our research team try to optimize the nitriding conditions to form a dense and sound CrN barrier by controlling various conditions. Thereafter, a diffusion couple test with a simulated TRU (Ce-Nd) was performed at 650 °C/25 hours to investigate the barrier performance.

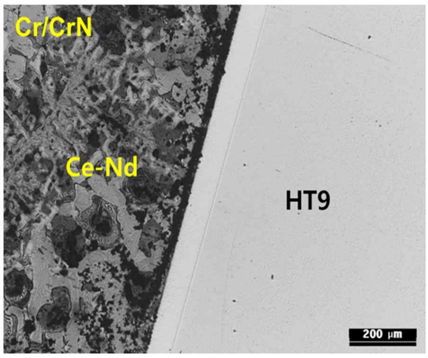

Fig.3. Result of the diffusion couple test between Cr/CrN barrier coating and Ce-Nd

Fig.3 shows the results of the diffusion couple test. FCCI was not observed even under more severe condition than actual nuclear power plants operating condition. From these result, it was confirmed that the CrN coating effectively inhibits the FCCI.

3. Suggestions for future research

Based on the research status regarding barrier coating technology introduced so far, the following three research topics are proposed. First, an optimization process of optimal conditions is required to form CrN coating stably and economically. Furthermore device improvement is essential to deposit uniform coating over the entire area of the long cladding tube of metric scale for practical use. Second, it is essential to evaluate the resistance to neutron irradiation as well as the inter-diffusion phenomenon due to high temperature. It is expected that the FCCI barrier performance will change according to the amount of damage to the barrier coating by neutrons. However, since the neutron irradiation test is difficult to access, an alternative test such as proton irradiation test is required. This alternative experiment can be utilized because it has been reported that irradiation defects and microstructure characteristics formed under appropriate proton irradiation conditions are very similar to those of neutron irradiation [10]. Finally, a fundamental analysis of the FCCI phenomenon is required. Although there are many studies related to FCCI, the mechanism is still unclear. The FCCI degradation mechanism which are changed with the materials composition and microstructures can be elucidated by systematic material engineering analysis. By using these results, modeling for FCCI degradation can be developed. This model can greatly help in material selection for next-generation nuclear power plants.

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