Structural Analysis of Surface-Modified Oxidation-Resistant Zirconium Alloy Cladding for Light Water Reactors

Youho Lee, Hee Cheon NO, and Jeong Ik Lee*
Dept. Nuclear & Quantum Engineering, KAIST, 373-1, Guseong-dong, Yuseong-gu, Daejeon, 305-701
**Corresponding Author: jeongiklee@kaist.ac.kr*
First Author: euo@kaist.ac.kr

1. Introduction

While the current zirconium-based alloy cladding (Zircaloy, here after) has served well for fission-product barrier and heat transfer medium for the nuclear fuel of light water reactors (LWRs) in steady-states, concerns surrounding its mechanical behavior during accidents have drawn serious attentions. In accidents, strength degradation of the current-zirconium based alloy cladding manifests at temperature around ~800ºC [1], which results in fuel ballooning. Above 1000ºC, zircaloy undergoes rapid oxidation with steam. Formation of brittle oxide (ZrO_2) and underlying oxygen-saturated α-zircaloy as a consequence of steam oxidation leads to loss of cladding ductility. Indeed, the loss of zircaloy ductility upon the steam oxidation has been taken as a measure of fuel failure criteria as stated in 10 CFR 50.46. In addition, zircaloy steam oxidation is an exothermic reaction, which is an energy source that sharply accelerates temperature increase under loss of coolant accidents, decreasing allowable coping time for loss of coolant accidents (LOCA) before significant fuel/core melting starts. Hydrogen generated as a result of zircaloy oxidation could cause an explosion if certain conditions are met. In steady-state operation, zircaloy embrittlement limits the burnup of the fuel rod to assure remaining cladding ductility to cope with accidents.

As a way to suppress hydrogen generation and cladding embrittlement by oxidation, ideas of cladding coating with an oxidation-preventive layer have emerged. Indeed, among a numbers of 'accident-tolerant-fuel (ATF)' concepts, the concept of coating the current cladding have drawn world-wide attentions as it is believed to be an effective - yet with low-risk-way of selectively suppressing the drawback of the current fuel rod [1-3]. Some signs of success on the lab-scale oxidation-prevention have been confirmed with a few coating candidates [4-6]. Yet, relatively less attention has been given to structural integrity of coated zirconium-based alloy cladding. It is important to note that oxidation-suppression performance of coated zirconium-based alloy cladding assumes mechanical integrity of the coating layer. Hence, maintaining mechanical integrity of coated layer as well as the underlying cladding material under realistic stress fields holds a key to success of coating technology. In this study, we investigate stress fields that coated zircaloy would experience during steady-state operation. With the obtained stress fields, we discuss mechanical integrity of coated zircaloy cladding with inferable failure modes.

2. Reference Coating Technologies

Al_3Ti-based alloys have been considered as a coating candidate in the hope of formation of preventive Al_2O_3 layer, which exhibit excellent oxidation resistance [7]. A research team at Korea Atomic Energy Research Institute (KAERI) showed that Al-21Ti-23 alloy and Al-25Ti-20Cr alloy exhibit an extremely low oxidation rate in a 1473K steam for up to 7200s when compared to zircaloy-4. Yet, the composition of Al_3Ti-based alloy is very important in its oxidation preventing capability; as an example the MAX phase coating – Ti_2AlC_2 – is found to be unprotective in steam-oxidizing environments [8]. Recently, KAERI identified two coating candidates – Silicon (Si) and Chromium (Cr) for future research concentrations to be made [4]. Both elements form oxidation-resistant oxide-scale with reasonable high temperature tolerance. KAERI investigated plasma spray (PS), and laser beam scanning (LBS) methods for coating Si and Cr onto the fuel cladding surface. They found that PS followed by LBS is more desirable to make a protective coating on the cladding surface [4]. The LBS treatment leads to a formation of mixed composition between Cr and Zr, identified as $ZrCr_2$. Indeed, Cr is also highly regarded as a potential cladding coating material in CEA [6]. CEA used physical vapor deposition (PVD) technology to coat Cr on the cladding surface. PVD coated Cr does not form a mixed composition with Zr, and it exhibited superior oxidation resistance over the Zircaloy-4 cladding for several hours exposure under steam at 1000ºC.

In this study, we select PVD Cr coating as a reference coating candidate as shown in Fig.1.

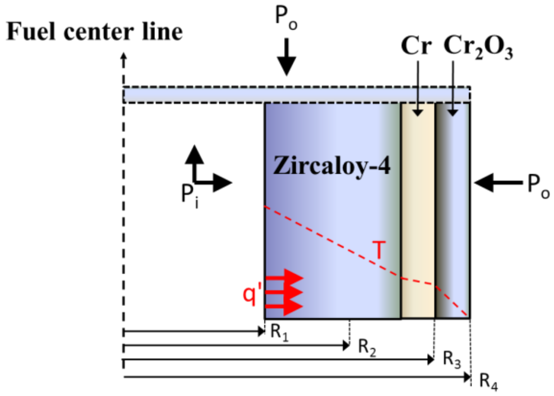

Fig. 1. Reference Chromium Coated Zr-4 Cladding

Different coating material may lead to different stress fields. Yet, given the zircaloy-4: metallic coating: oxide scale structure remains, the general structural behavior and integrity assessment would remain the same. Total cladding thickness of standard 0.57mm was used, 80%, 10%, and 10% of the total cladding thicknesses were assumed to be Zr-4, Cr, and Cr_2O_3, respectively, as illustrated in Fig.1. Coating processes result in a formation of a distinct coating layer. Upon the oxidation of the coating layer, a distinct oxide layer is formed on the coating surface. Hence, we essentially treat the coated zirconium-based alloy as a triple-laminated cylindrical layers that are constrained with strain compatibility at the interfaces. The following material properties were used.

Table I: Reference Material Properties for Cr-Coated Zr-4 Cladding Stress Analysis

	Zr-4	Cr	Cr_2O_3
Elastic Modulus [GPa]	68	279	230
Poisson Ratio [-]	0.3	0.21	0.25
Thermal Conductivity [W/m-K]	16.4	80.0	3.18
Thermal Expansion Ratio [1/K]	$6.6\text{x}10^{-6}$	$8.45\text{x}10^{-6}$	$6.5\text{x}10^{-6}$

3. Multi-Layer Cladding Structural Analysis Code

The multi-layer cladding stress analysis code developed by the first author at Massachusetts Institute of Technology (MIT) [9] was used. The code calculates stresses arising from mechanical and thermal loading in principal directions using linear-elasticity. Fig.2 shows the comparison between ANSYS-obtained multi-layer cladding stress fields and ones obtained by MIT structural analysis code for the same reference Cr-coated cladding geometry and operating conditions.

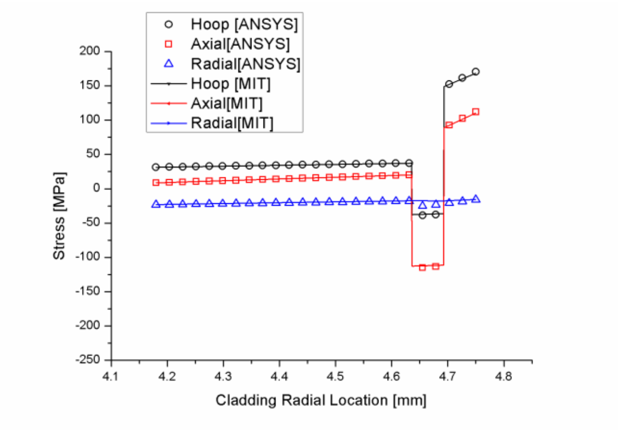

Fig. 2. ANSYS-Mechanical validation of Multi-layer cladding structural model for surface modified zirconium based cladding [P_i=23MPa, P_o=15.5MPa, q'=18kW/m]

We confirmed that the MIT structural code gives a good agreement with FEM obtained stress fields for coated cladding.

4. Burnup-Dependent Cladding Structural Integrity

Cladding experiences different stress distributions with incore residence time. That is, fuel rod internal pressure build-up with burnup, pellet-cladding mechanical interaction (PCMI), and cladding material property changes with irradiation, and corrosion. In addition, fuel pin power changes with fuel shuffling and depletion. Among those factors, cladding stress level is the most sensitively affected by the degree of gap interfacial pressure arising from burnup-dependent PCMI; cladding experiences an internal pressure swing ranging 5 ~ 25 MPa during its incore residence time, depending on the degree of fission gas release, and PCMI. Although the other factors would also affect cladding stress level evolutions, they seem quite marginal compared to such a dramatic internal pressure swing. Hence, for the purpose of preliminary assessments, we consider burnup-dependent cladding structural integrity in terms of different internal pressurization.

Different cladding layer properties, yet with strain compatibility at the interfaces, lead to hoop (tangential), and axial stress discontinuities in the radial direction, as shown in Fig.3. A noticeable feature is that in the low burnup, the Cr coating layer is under high compressive stress. With increasing burnup that gives a higher internal pressure, the stress level in Cr coating layer relieves whereas the protective oxide layer sees increasing tensile stresses.

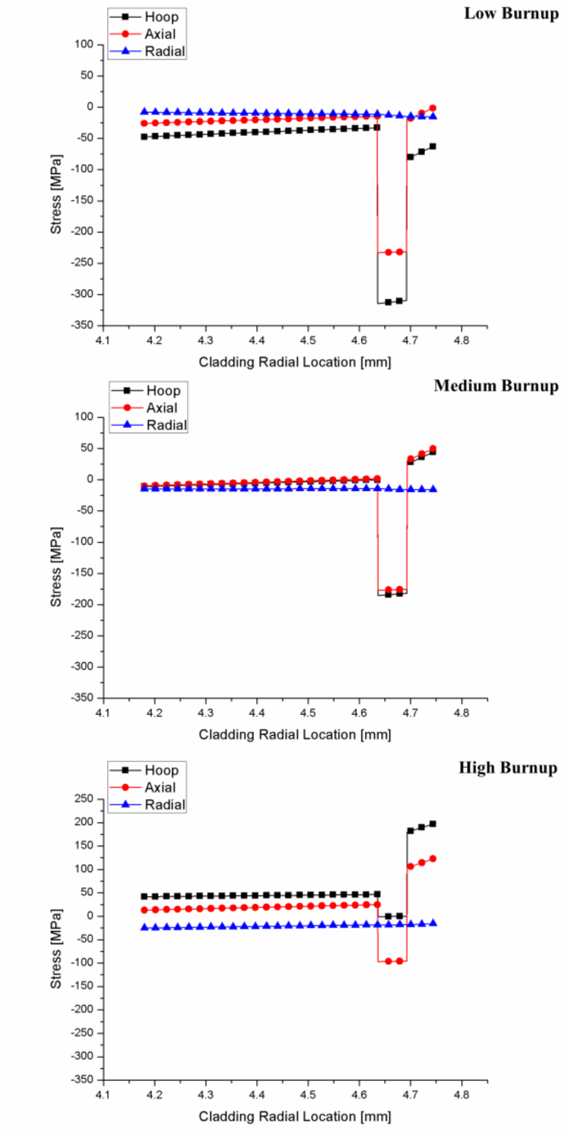

Fig. 3. Burnup-dependent Cr-coated zircaloy-4 cladding stress distribution [P_i=8MPa, 15MPa, and 25 MPa for low, medium, and high burnup, respectively, P_o=15.5MPa, q'=18kW/m]

Such bunrup-dependent cladding stress evolution implies time-dependent failure modes for surface modified zirconium based alloy cladding. In the low burnup, if failure occurs, it is likely to be the coating material that is placed between the zircaloy and the oxide layer. The local plasticity may occur in the coating layer and the resulting strain may lead to mechanical deformation of the coating layer. Yet, compressive stresses are anticipated not to cause fractures in the oxide layer in the low burnup operations as long as the underlying coating layer remains intact. As shown in Fig. 4, the chromium layer sees the highest von Mises stresses during the low burnup period due to high compressive stresses.

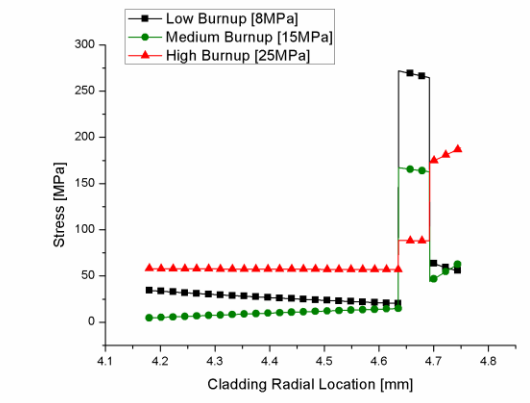

Fig. 4. Burnup-dependent von Mises Stress for Cr-coated zircaloy-4 cladding [P_o=15.5MPa, q'=18kW/m]

From the view point of metallic failure that is often defined as onset of yielding, compressive stresses equally contribute to a failure in comparison with tensile stresses. Hence, from the view point of the Cr layer mechanical integrity, high compressive stresses during the low burnup operation is detrimental. However, for the outer oxide layer, which essentially exhibit brittle behavior, tensile stresses are far more dangerous than compressive stresses. Indeed, such ceramic materials exhibit usually a few times lower fracture stress under tension than under compression. Hence, particular attention should be given to the integrity of the external oxide coating layer for high burnup situations. We can see that the oxide layer experiences as high as ~200 MPa hoop stress in high burnup. Leaving large ceramic structure under the ~200MPa of tensile stresses is certainly beyond the norm of the material usage. While the brittle oxide layer would exhibit statistical fracture modes under applied stresses, one can strongly suspect dispersed fractures for a thin layer of 3.8m long ceramic material under tensile stresses of ~200 MPa (hoop), and ~100 MPa (axial), in their principal directions.

5. Conclusions: a Key Issue Identified

From the stress analysis of the coated zirconium-based alloy cladding with Cr, we would like to inform ATF communities that there could be potentially a structural issue for high burnup operation of coated zircaloy cladding. We anticipate noticeable dispersed fractures of the protective oxide layer once fuel rod reaches an interfacial gap pressure – not particularly higher than the typical gap interfacial pressure that we would normally expect as a result of pellet and cladding mechanical interaction (PCMI). Hence, in order for the cladding coating idea to survive and gain much more confidence, we recommend experimentalists to run their oxidation experiments under high-burnup simulating stress fields. A material that could sustain its mechanical integrity under such simulated stress fields should be treated as a potential coating candidate. This study may be used to infer a key technical challenge associated with cladding surface modification concept – that one may be able to understand with a mere common sense; in notorious incore environments, laminating multiple layers would be never as easy as one can easily expect in the lab.

REFERENCES

[1] K. Barret, S. Bragg-Sitton, and D. Galicki, Advanced LWR Nuclear Fuel Cladding System Development Trade-off Study, Idaho National Laboratory, 2012.

[2] A. M. Yacout, M. Pellin, and M. C. Billone, Development and Testing of Nanolaminate Coatings for Conventional LWR Cladding, TopFuel, Charlotte, North Carolina, September 15-19, 2013.

[3] AREVA Federal Services, LLC, Interim Analysis Report for Enhanced Accident Tolerant Fuels, AREVA, 2013.

[4] Y-H. Koo, J-H. Yang, and J-Y. Park, KAERI's Development of LWR Accident Tolerant Fuel, Journal of Nuclear Materials, submitted, under review, 2013.

[5] H-G. Kim, et.al., High-Temperature Oxidation Behavior of Cr-Coated Zirconium Alloy, TopFuel, Charlotte, North Carolina, September 15-19, 2013.

[6] I. Idarraga-Trujillo, et.al., Assessment at CEA of Coated Nuclear Fuel Cladding for LWRs with Increased Margins in LOCA and Beyond LOCA Conditions, TopFuel, Charlotte, North Carolina, September 15-19, 2013.

[7] J-Y. Park, High Temperature Steam Oxidation of Al_3Ti-based Alloys for the Oxidation-Resistant Surface Layer on Zr Fuel Claddings , Journal of Nuclear Materials, Vol. 437, pp. 75-80, 2013.

[8] Personal correspondence, Center for Advanced Nuclear Energy Systems, Massachusetts Institute of Technology, Cambridge, U.S.A, 2013

[9] Y. Lee, and M. S. Kazimi, A Structural Model for Multi-Layerd Ceramic Cylinders and its Application to Silicon Carbide Cladding for Light Water Reactor Fuel, Journal of Nuclear Materials, Submitted, under review, 2014.