Initial Thermal-Hydraulic Regulatory Points on Accident Tolerant Fuel Design Review

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1. Introduction

After Fukushima Daiichi nuclear power plants accident in 2011, Accident Tolerant Fuels (ATFs) aiming at improved integrity under accidents conditions compared to the conventional nuclear fuels with UO_2 pellet and zircaloy alloy cladding have been actively developed worldwide. In Korea, to keep pace with this move, an ATF having the chromium coated HANA-6 cladding and the UO_2 pellet doped with La_2O_3-Al_2O_3-SiO_2 is currently being developed by Korean nuclear industry and it is scheduled to be submitted to Korea Institute of Nuclear Safety (KINS) as a topical report application by 2029. According to this Korean nuclear industry move, KINS is developing a regulatory guide on ATF design review by implementing the research program titled as "Study on Validation of the Consolidated Safety Analysis Platform for Applications of Enhanced Safety Criteria and New Nuclear Fuels" since 2021.

Although any topical report for a new fuel design usually highlights specific evaluations results on fuel rod and fuel assembly, it should also include the impact analysis of the new fuel design on reactor physics, core thermal-hydraulic, and accident analyses. Therefore, in the present study, we narrow down our focus on the accident analyses and suggest several initial regulatory points which can be applied to ATF design review process from thermal-hydraulic point of view.

2. Identification of Initial Regulatory Review Points from Thermal-Hydraulic Point of View

Research results from domestic and international investigations on ATFs have been complied to identify the initial thermal-hydraulic regulatory review points. Especially, safety analysis codes related issues are mainly focused. In the present study, the MARS-KS Thermal-Hydraulic (TH) system code is taken into account because the MARS-KS is the audit code used by KINS and it has also wide user groups such as academia, research institutes and nuclear industry in Korea.

2.1 Ability to Model a Multi-Layer Cladding Structure

Since the most common design basis accident usually analyzed for the safety analysis with a TH system code is Large Break Loss Of Coolant Accident (LB LOCA), an illustrative LB LOCA analysis was conducted to identify any impact of a multi-layer cladding structure of a typical ATF such as the chromium coated zircaloy alloy cladding. [1-3]

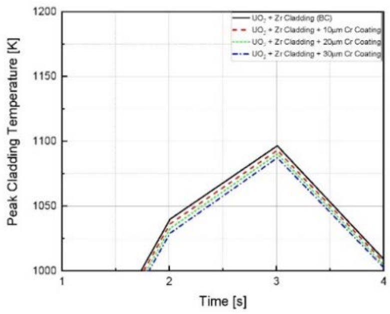
The MARS-KS input of Zion power plant for OECD-BEMUSE Phase IV project has used for this illustrative analysis. Here, two approaches were adopted. One approach is to introduce equivalent thermal properties for thermal conductivity and thermal capacity in order not to change the MARS-KS code (i.e. keeping the original zircaloy metal-water reaction oxidation model). The other approach is to modify the MARS-KS code coding to accommodate a multi-layer structure by chromium coating including a chromium-based metal-water reaction oxidation model. Analyses have been done for three chromium coating thicknesses and the results of both approaches were compared with that of the base case without chromium coating. Table I shows the Peak Cladding Temperature (PCT) of Zion power plant LB LOCA analysis with the MARS-KS code. [2]:

Table I: Comparison of PCTs for Zion power plant LB LOCA

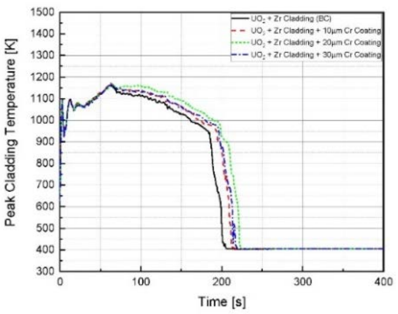
Peak Cladding Temperature (PCT, Kelvin)					
Approach	Phase	Zr-4(Base)	10μm Cr coating	20μm Cr coating	30μm Cr coating
Equivalent thermal properties	Blowdown	1096.5	1093.0	1090.2	1087.2
	Reflood	1163.4	1171.2	1169.6	1165.4
Multi-layer structure	Blowdown	1096.5	1093.5	1090.5	1087.8
	Reflood	1163.4	1168.6	1182.6	1166.9

Table I shows that for the blowdown phase, PCTs are decreasing as chromium coating thickness increases and they remain lower than that of base case. Since the blowdown PCTs are well matched for the two approaches employing different metal-water reaction models and blowdown peaks appear almost immediately after the LOCA initiation, the metal-water reaction does not seem to play much for the blowdown phase. Considering much higher thermal conductivity of chromium (around 4 times compared to Zr-4) and thin chromium coating thickness from 10μm to 30μm compared to Zr-4 cladding thickness (0.61mm), equivalent thermal conductivities of the chromium coated Zr-4 cladding increase from 1% to 3% compared to original Zr-4 cladding. [3] By the way, it is well known that the initial overheating of the cladding during LB LOCA is due to redistribution of the heat stored on the fuel rods for normal operation [4,5]. Therefore, considering the fact that initial stored energy of fuel rod during a normal operation might be lower as the chromium thickness gets thicker due to the increase of equivalent conductivity of the cladding and the heat transfer area, the decreasing trend in PCT for the blowdown phase seems to be the thickening effect of chromium coating.

For the reflood phase, Table I does not show any clear trend in PCTs with chromium thickness except that PCTs from all chromium coating thicknesses for both approaches remain higher than that of base case. Especially, the maximum increase of PCT even amounts to about 20 Kelvin for the multi-layer structure approach (for 20µm Cr coating case). The reflood PCTs of the equivalent thermal properties approach employing the original zircaloy metal-water reaction show lower PCTs for 20µm and 30µm chromium coating cases compared to those of the multi-layer structure approach. Therefore, the metal-water reaction doesn't seem to play an important role in deciding PCTs in the reflood phase. Since the reflood PCT is known to be strongly sensitive to the dispersed film boiling heat transfer [6,7] and the MARS-KS code's dispersed film boiling heat transfer model has T_{wall} in its formulation [8], it seems that all increases of PCTs in the reflood phase in Table 1 are due to the degradation of the dispersed film boiling heat transfer by T_{wall} change because of effective cladding conductivity change by chromium coating. For reference, LB LOCA calculation result for Zion plant conducted by the equivalent thermal properties method is shown in Figure 1 below.



(a) Blowdown phase PCT behavior



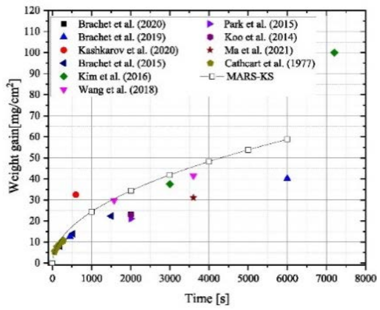
(b) Overall PCT behavior

Fig. 1 PCT behavior depending on chromium coating thickness by adopting the equivalent thermal properties method. [1,3]

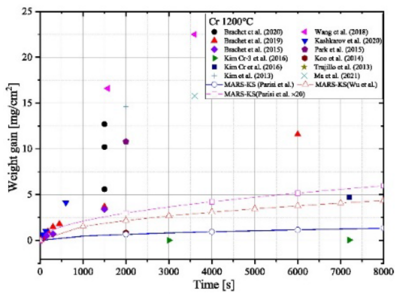
From these observations, it is obvious that chromium coating changes the blowdown and the reflood PCTs of LB LOCA. Therefore, a TH system code used for safety analysis of an ATF having the chromium coating should have capacity to simulate the multi-layer feature properly for reliable calculation of PCT for LB LOCA.

2.2 Ability to Model High Temperature Oxidation of a Chromium-coated Layer

It is known that the high temperature oxidation model included in the MARS-KS code is developed from Cathcart-Powel's zircaloy based oxidation model. In addition, most of ATFs being developed these days adopt a chromium coating on zircaloy alloy cladding of the primary coolant side to take advantage of the high temperature oxidation resistant property of chromium. Otherwise, the other face of zircaloy alloy cladding normally does not have any chromium coating on it. Therefore, it is obvious that a chromium based high temperature oxidation model should be added to evaluate double sides oxidation of ATF during LB LOCA correctly.



(a) Zircaloy



(b) Chromium

Fig. 2 Comparison of Weight Gain of High Temperature Oxidation for Zircaloy and Chromium at 1200℃.[9]

Recent study regarding chromium-based oxidation models clearly shows that high temperature oxidation rates between chromium and zircaloy are quite different from each other. [9] Figure 2 shows that weight gains of zircaloy and chromium at 1200℃ are quite different.

Therefore, a TH system code used for safety analysis of an ATF having the chromium coating on the primary coolant side should have additional capacity to simulate the high temperature oxidation of chromium properly for reliable calculation of Equivalent Cladding Reacted (ECR) rate for LB LOCA.

2.3 Effect of Chromium Coating on Critical Heat Flux and Minimum Film Boiling Temperature

For When LB LOCA occurs, complex thermal-hydraulic phenomena happen due to rapid depressurization and voiding of the reactor core as the accident progresses from blowdown, through refill, up to reflood phases. Critical Heat Flux (CHF) generating Departure of Nuclear Boiling (DNB) is also a phenomenon belonging to these complex phenomena. Since it is believed that CHF depends on surface conditions such as 1) roughness, 2) wettability, and 3) porosity (of a crud layer) [10, 11], many CHF experiments on chromium-coated fuels have been conducted. Unfortunately, however, it is not crystal clear that chromium coating really helps increase or decrease CHF until now [3] and because of that, the US NRC recommends that a reviewer of thermal and hydraulic design section should ensure that applicants appropriately address the following areas with justification for PWRs [11]

- Whether changes to hydraulic diameter due to the coating thickness affect the applicability of the CHF or CP correlation
- Whether the addition of a chromium coating, including consideration of the effects of surface roughness, changes the fuel rod boiling crisis behavior

Some researchers argued that flow regime inside the reactor core is “Flow Boiling” and as a result, the surface condition might not have much effect on CHF as “Pool Boiling” case where the surface condition might have much effect on CHF. [12] However, considering the inside core during the LOCA period, the condition seems far from “Flow Boiling” and close to “Pool Boiling” given the reflooding fluid rises very slowly during the reflood phase. For a typical 25.4mm/sec, the reflood velocity corresponds to 25.4kg/m^2-sec (when density of water is assumed 1,000kg/m^3) and this value is much lower than 200kg/m^2-sec where they found that 50% difference in CHF appears among various cladding materials. [12] Therefore, it is safe to assume that CHF can be different depending on cladding materials specially during the reflood period of LB LOCA. This observation that the CHF could change with the cladding materials close to the pool boiling condition is compatible with the recent study [13] where the pool boiling CHF correlation of Kandlikar [14] (Equation (1)) having the contact angle dependency was confirmed by physical experiments.

$$q_{CHF} = h_{lg}\rho_g^{0.5}\left[\sigma g(\rho_l-\rho_g)\right]^{0.25}\left(\frac{1+\cos\theta_r}{16}\right)\left[\frac{2}{\pi}+\frac{4}{\pi}(1+\cos\theta_r)\cos\phi\right]^{0.5} \quad (1)$$

where h_{lg} is the water latent heat, ρ_g and ρ_l are the steam and water density,σ is the water surface tension, g is the acceleration of the gravity, θ_r is the receding contact angle, ϕ is the surface inclination angle.

Regarding the heating surface effect on the minimum film boiling temperature (MFBT) (or Leidenfrost temperature, Rewet temperature), as noted by Hong [3], Ghiaasiaan [15] pointed out that the heating surface property could affect Leidenfrost temperature of the pool boiling with reference to Henry’s research. [16] In fact Henry’s Leidenfrost temperature model was developed by correcting Berensen’s Leidenfrost temperature model [17] taking into account the transient conjugate heat transfer in the surface.

$$T_{MFB,Berenson} = T_{sat} + 0.127\frac{\rho_{g,film}}{k_{g,film}}\left[\frac{g(\rho_l-\rho_g)}{(\rho_l+\rho_g)}\right]^{2/3}\left[\frac{\sigma}{g(\rho_l-\rho_g)}\right]^{1/2}\left[\frac{\mu_{g,film}}{g(\rho_l-\rho_g)}\right]^{1/3} \quad (2)$$

$$T_{MFB,Henry} = T_{MFB,Berenson} + 0.42(T_{MFB,Berenson} - T_l)\left\{\left[\frac{(\rho k c_p)_l}{(\rho k c_p)_{wall}}\right]^{1/2}\left[\frac{h_{lg}}{c_{p,wall}(T_{MFB,Berenson}-T_{sat})}\right]\right\}^{0.6} \quad (3)$$

where T_{sat} is the saturation temperature, $k_{g,film}$ is the thermal conductivity of steam at the film temperature condition, $\rho_{g,film}$ is the density of steam at the film temperature condition, $\mu_{g,film}$ is the viscosity of steam at the film temperature condition, $c_{p,wall}$ is the specific heat at constant pressure of wall, k_{wall} is the thermal conductivity of wall, $c_{p,l}$ is the specific heat at constant pressure of water, k_l is the thermal conductivity of water.

Looking into Eqn. (2) and (3), one can notice that there are some wall related properties inside Henry’s model otherwise there is nothing related to wall properties inside Berenson’s model. For reference, the MARS-KS code adopts Henry’s model but other thermal-hydraulic codes may not adopt the same model for their calculation of Leidenfrost temperature. There are many other studies regarding the surface effect on MFBT and one of them [18] shows that the porosity could have deep impact on Leidenfrost temperature. Recently, Seshadri et al.[19] found that Leidenfrost temperature becomes lower and quenching speed becomes slower for the chromium coated zircaloy cladding compared to the zircaloy cladding without the chromium coating. They also found that these trends in Leidenfrost temperature and the quenching speed becomes reversed for irradiated cladding cases. Table II shows their measurements of Leidenfrost temperature and quench front speed.

Table II: Quench Results [19]

Sample	Leidenfrost temperature (℃)	Quench front speed (mm/s)

Non-irradiated samples		
Zirc-4	302	9.6
Cr Coated	293	6.8
Gamma irradiated samples		
Zirc-4	315	10.6
Cr Coated	323	12.5

The US NRC also recommends that a reviewer of a coated cladding should ensure whether the addition of a chromium coating affects the rewet temperature following dryout for BWRs.[11] All those observations implies MFBT (Leidenfrost temperature) becomes different depending on cladding materials type and its surface condition. Since CHF and MFBT are factors which impact PCT when LB LOCA is analyzed by a TH system code, it is advisable that chromium coating effect on CHF and MFBT should be modeled precisely into the TH system code or extra uncertainties from the chromium coating to CHF and MFBT should be considered during LB LOCA Best Estimate Plus Uncertainty (BEPU) calculation.

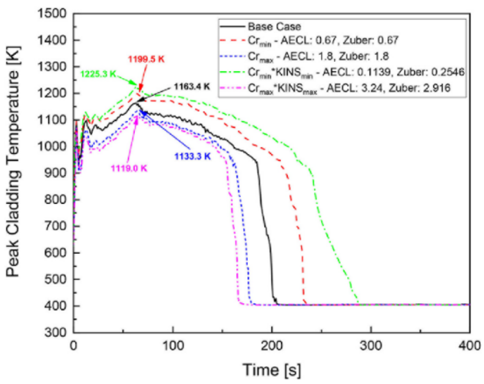


Fig. 3 PCT Variations depending on CHF Changes [1]

For reference, a LB LOCA calculation result for Zion plant is presented in figure 3 where the original CHF models of the MARS-KS code has intentionally adjusted incorporating observed CHF variation of chromium coated zircaloy claddings over that of conventional zircaloy claddings [67%($KINS_{min}$)~180%($KINS_{max}$)). [1] Note that the MARS-KS code is employing Groeneveld (AECL) CHF lookup table for the blowdown phase with an uncertainty range of 0.17~1.8 and Zuber CHF correlation for the reflood phase with an uncertainty range of 0.38~1.62.

2.4 Effect of Chromium Coating on Film Boiling and Transition Boiling Heat Transfer

Iloeje [20] asserted that the post-dryout heat transfer is compose of three heat transfer mechanisms and the two mechanisms by droplet collision with the wall and the wall-gas-droplet sequential heat transfer except the heat transfer between the wall and gas are affected by wall surface conditions. Brown et al. [21] showed that film boiling heat transfer coefficient measured by University of New Mexico for the flow boiling experiment with FeCrAl cladding amounts to 30~100% of RELAP5-3D code-calculated film boiling heat transfer coefficient. He et al.[22] pointed out that the thermal-physical properties such as volumetric heat capacity, thermal conductivity, and the thermal emissivity of a cladding affect the film boiling heat transfer. For reference, the film boiling heat transfer model of the MARS-KS code for LB LOCA reflood phase is given by

$$q''_{film} = q''_{Bromley} + q''_{Rad} \qquad (4)$$

$$q''_{Bromley} = 0.62\left(\frac{D_H}{\lambda_c}\right)^{0.172}\left[\frac{k_g^3\rho_g(\rho_l-\rho_g)h'_{lg}g}{D_H\mu_g(T_w-T_l)}\right]^{1/4}(T_w-T_l)$$

$$q''_{Rad} = \frac{\sigma_{SB}}{\frac{1}{\varepsilon}+\frac{1}{\alpha_L}-1}(T_w^4-T_l^4) \text{ and } \lambda_c = 2\pi\left[\frac{g_c\sigma}{g(\rho_l-\rho_g)}\right]^{1/2}$$

where $q''_{Bromley}$ is the modified Bromley correlation representing the convection part of the film boiling heat transfer, D_H is the hydraulic diameter, g_c is the gravitational constant, q''_{Rad} is the radiation part of the film boiling heat transfer, h'_{lg} the enthalpy difference between vapor and saturated water, ε is the thermal emissivity of cladding, α_L is the absorptivity of liquid, σ_{SB} is Boltzman constant.

Through Eq. (4), one can notice that at least T_w and ε reflects the cladding material properties effect on the film boiling heat transfer.

In the previous section, it is shown that CHF and MFBT of the chromium coated zircaloy alloy cladding could be different from those of the conventional zircaloy alloy claddings. This observation leads to an educated guess that the transition boiling curve of the chromium coated zircaloy alloy might be also different from those of the conventional zircaloy alloy claddings because the transition boiling curve could be obtained by the interpolation between CHF and MFBT points. Since the film and the transition boiling are factors affecting PCT when LB LOCA is analyzed by a TH system code, it is advisable that chromium coating effect on the film

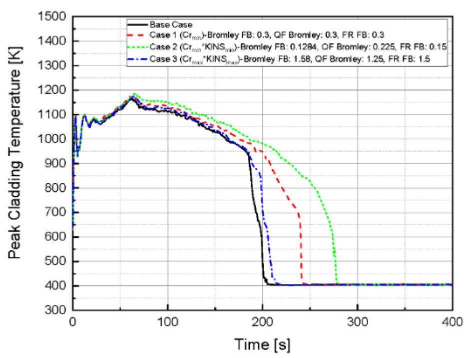


Fig. 4 PCT Variations depending on Film Boiling Heat Transfer Change [3]

and the transition boiling should be reflected precisely into the TH system code or extra uncertainties due to the chromium coating should be incorporated into the LB LOCA BEPU calculation process. For reference, LB LOCA calculation result for Zion plant is presented in figure 4 where the original film boiling correlations of the MARS-KS code has intentionally adjusted incorporating observed maximum film boiling heat transfer reduction of FeCrAl claddings (30%). [22] Note that the MARS-KS code is employing Bromley film boiling correlation with an uncertainty range of 0.428~1.58, Quench Front Bromely correlation (for the reflood phase) with an uncertainty range of 0.75~1.25, and Forslund-Rohsenow film boiling correlation (for the reflood phase) with an uncertain range of 0.5~1.5.

2.5 Effect of Chromium Coating on Nucleate Boiling Heat Transfer

There are not many researches on chromium coating effect on nucleate boiling heat transfer. However, Lee et al. [12] shows that surface characteristics and material thermal effusivity (conductivity and capacity) have a clear relationship to nucleate boiling heat transfer behavior for the flow boiling where mass flow is low.

In terms of nucleate pool boiling, Rohsenow empirical correlation [23] is widely used and the heat transfer coefficient of Rohsenow is given by

$$h_{Rohsenow} = \mu_l h_{lg} \left[\frac{g(\rho_l-\rho_g)}{\sigma}\right]^{0.5} \left[\frac{c_{p,l}}{C_{s,l} h_{lg} Pr_l}\right]^3 (T_w - T_{sat})^2 \qquad (5)$$

where, μ_l is the water viscosity, Pr_l is the Prandtl number of water, and $C_{s,l}$ is the model coefficient specific for a fluid-surface combination. (for example, 0.0133 for water-stainless steel).

Pioro [24] showed $C_{s,l}$ can be different for different combinations of fluids and surface materials and suggested 0.019 for water/chromium combination (See, Table I of Pioro [24])

Kurul and Podowski [25] suggested a mechanistic wall boiling model for nucleate pool boiling and they partitioned heat flux from heated wall into three components, 1) single-phase convective heat flux, 2) heat flux associated with phase change (evaporation), 3) quenching heat flux, transferred to the liquid phase during the wait time. Here, the wait time means the time to complete the process by which cooler liquid fills space near heated wall vacated by the departing bubble, thermal boundary layer is rebuilt, nucleation of the next bubble occurs when the critical superheat condition in the cavity is reached. Therefore, the wait time (especially a time to build the thermal boundary layer again) is likely to be affected by transient behavior of wall temperature. Since a surface material effusivity can impact the transient behavior of the wall, it means that quenching heat flux and as a result the nucleate pool boiling heat transfer may change depending on the heating wall material.

From these observations, it seems reasonable to assume that the chromium coating on the zircaloy cladding has some impacts on the nucleate pool boiling and the nucleate flow boiling with low mass flow rate and it affects the reflood phase of LB LOCA calculation.

Therefore, it is advisable that chromium coating effect on the nucleate boiling should be reflected precisely into the TH system code or extra uncertainties due to the chromium coating should be incorporated into the LB LOCA BEPU calculation process. For reference, the MARS-KS code employs Chen nucleate boiling model. [26]

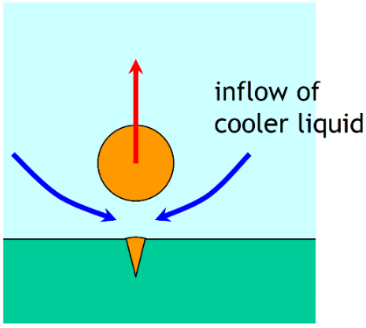


Fig. 5 Bubble behavior during wait time

2.6 Impact on Thermal-Hydraulic Characteristics by Surface Condition Changes due to Chromium Coating Damages

Due to the difference in irradiation growth rate between chromium and zircaloy, tensile stress is exerted at the interface of two materials. As a result, chromium coating might have cracking or delamination during the normal operation. In addition, if LB LOCA occurs, fuel rods would get ballooned and this ballooning may impose large strain which might accelerate cracking or delamination of the chromium coating. In fact, the PIRT by the US NRC ranked coating cracking and delamination as “high” during accident conditions. [11]

Since cracking or delamination of the chromium coating layer means a significant change in “Surface condition” and this could lead to changes of thermal-hydraulic characteristics such as CHF, MFBT, film boiling, transition boiling, nucleate boiling etc., the US NRC recommend that the reviewer of Final Safety Analysis Report, Section 4.4 (Thermal and Hydraulic Design) should ensure that these damages are appropriately accounted for or that coating degradation is otherwise prevented. [11]

Therefore, unless the chromium coating’s integrity is guaranteed under the design basis accident conditions where the coating integrity may be jeopardized the most (e.g. LB LOCA, RIA), the surface condition change from coating cracking and delamination should be properly reflected into thermal-hydraulic characteristics of a TH system code.

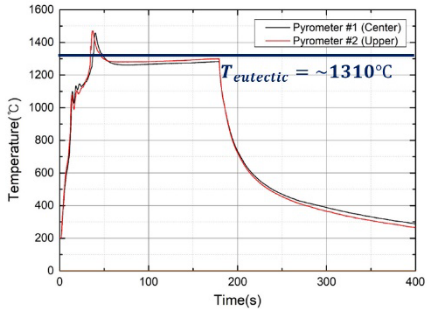
2.7 Impact on Thermal-Hydraulic Characteristics by Surface Condition Changes due to Eutectic Formation

It is well known that the eutectic formation exists at the interface between chromium coating and zircaloy alloy substrate. The eutectic formation is a phenomenon that the melting temperature at the interface becomes lower than the original melting temperatures of each of material formed a cladding. In fact, it is reported that the lowest eutectic formation temperature for the chromium-zircaloy system is about 1,332℃ despite the fact that the melting temperatures of chromium and zircaloy are 1,857℃ and 1,852℃, respectively. [10]

One of distinctive features of the eutectic formation is that the surface condition can change drastically within short period of time. Figure 6 shows a very rough surface is formed by the eutectic phenomena within a few tens of seconds. [27] Unlike a normal zircaloy cladding which has no significant change in surface morphology, there is a remarkable change in surface morphology for the chromium coated zircaloy cladding when the cladding temperature becomes higher than the eutectic temperature. This highly uneven surface pattern is called as “crocodile skin shape”. [28]

An ATF, by its definition, should be more resilient for Beyond Design Basis Accidents (BDBA) conditions compared to conventional zircaloy fuels. Therefore, its better performance under BDBAs conditions should be demonstrated when the ATF application is submitted to a regulatory body. Unfortunately, however, it is highly likely that in case of BDBAs conditions, fuel cladding temperature exceeds the eutectic temperature as the accident develops and the cladding surface turns to the crocodile skin shape. What this transition of the surface shape implies is that a TH system code to analyze BDBAs for the chromium coated zircaloy cladding should have capacity to accommodate the change of thermal-hydraulic characteristics due to the surface morphology variation by the eutectic formation so that it can produce reliable analyses for BDBAs.

(a) Formation of Crocodile Skin Shape



(b) Heating History for the Experiment

Fig. 6. The Eutectic Phenomena in Chromium-Zircaloy-1.1Nb System [27]

3. Conclusions

In the present study, international and domestic researches on ATF have been compiled to identify initial regulatory review points for ATF design review on the safety analyses area from the thermal-hydraulic perspective.

Through the in-depth literature review, a total of 7 initial regulatory review points are identified. KINS is applying the present results to the ATF LTR (Lead Test Rod) and HANA-6 cladding topical report applications reviews in progress, and the review focuses are going to apply for LTA (Lead Test Assembly) and ATF topical report applications reviews in the future.

Some of the review focus items may require an applicant long-term and time-consuming preparation or study for their substantiation. Therefore, the applicant may want to consult the present result to set up specific items for their long-term research and development plan for their ATF project.

In closing, it should be noted that the regulatory review focuses presented here just represents preliminary level of regulatory points based on current knowledge level. Therefore, the possibility to emerge additional regulatory review focuses or update some of the present result is still open

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