Transactions of the Korean Nuclear Society Autumn Meeting
Gyeongju, Korea, October 26-27, 2017

Corrosion Related Unidentified Deposit Adhesion Test and Analysis in Simulated Pressurized Water Reactor Conditions

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1. Introduction

As commercial nuclear power plant, has been operated for a long term, there are some unwanted deposit on the upper side of cladding surface, which was observed from Callaway pressurized water reactor (PWR) in United States during the 9th cycle.

This kind of problem is caused by combination of heat transfer and corrosion phenomenon. According to coolant flow direction through the fuel assembly, surface temperature of cladding at the upper side is higher than that of lower side. When surface temperature rises over saturation temperature of the coolant, sub-cooled boiling can occur on that point and this can form porous corrosion related unidentified deposit (CRUD) on it.

CRUD can cause several problems during the normal operation period. For example, if boron, inside of coolant, enters into the CRUD, this compound will absorb neutron a lot which causes severe problem related with neutron flux named axial offset anomaly (AOA) or CRUD induced power shift (CIPS)

And also, CRUD may cause a problem concerned with corrosion mechanism. Since the characteristic of CRUD is porous as abovementioned, some corrosive solution can permeate into the pore of CRUD. If it stays for a long time, CRUD induced localized corrosion (CILC) problem occurs.

In this point of view, investigating CRUD should be conducted to solve the problems AOA and CILC. Following this motivation, we were planning to construct CRUD adhesion test equipment for various research related with CRUD. In this paper, the equipment set-up logics will be explained under heat transfer, thermohydraulic and CRUD adhesion test and analysis result will be introduced.

2. Methods and Results

In this section, the test equipment set-up logistics for CRUD adhesion test will be introduced. The test environmental conditions were set by following several heat transfer and thermohydraulic calculations which can make sub-cooled boiling phenomenon on the sample surface.

2.1 Sample Surface Temperature

To simulate CRUD adhesion on the sample surface, generating sub-cooled boiling on it is the most important, which has been known as the main mechanism of CRUD adhesion on the cladding surface.

Sub-cooled boiling phenomenon occurs when heated surface temperature becomes higher than saturation temperature of liquid whose bulk temperature is lower than that. In this situation, some bubbles are generated on the heated surface, and this process continues until phase change of boiling to film boiling.

In this study, the basic water chemical conditions are followed as that of primary circuit environment of commercial nuclear power plant; pressure is 15.5 MPa. Therefore, we should set the temperature of sample surface over the saturation temperature of coolant in 15.5 MPa environment.

Refer to thermodynamics properties table, 1st order extrapolation was conducted to calculate the saturation temperature of water in 15.5 MPa. The result of this calculation fits with that of real condition in primary circuit water chemistry of PWR, 345 ℃.

To set the exact temperature of sample surface, sub-cooled boiling conditions in PWR and permissible range of cladding surface temperature should be considered. According to text book of Neil E et al. [1], sub-cooled boiling starts occurring when the cladding surface temperature becomes higher about 0.1 ℃ more than the saturation temperature of coolant, 345 ℃. And IAEA sets regulation of cladding surface temperature as 347 ℃ [2].

Following these considerations, we give engineering margin from the minimum temperature to generate sub-cooled boiling but not to be exceeded the limitation. Therefore, our test condition of surface temperature was set as 346 ℃.

2.2 Heat Flux for Sub-cooled Boiling

By using the surface temperature set from previous section, we can estimate how much heat flux is needed on the sample surface.

The heat flux for sub-cooled boiling can be calculated by using the empirical equation suggested by Rohsenow et al. in 1952 [3]. According to his research, heat flux to generate sub-cooled boiling can be written as below equation 1.

$$\dot{q}_{nucleate} = \mu_l h_{fg}\left[\frac{g(\rho_l-\rho_v)}{\sigma}\right]^{\frac{1}{2}}\left[\frac{C_{pl}(T_{sur}-T_{sat})}{C_{sf}h_{fg}Pr_l^n}\right]^3 \quad (1)$$

$\dot{q} = heat\ flux$
$\mu = dynamic\ viscosity$
$h_{fg} = enthalpy\ of\ vaporization$
$g = acceleration\ of\ gravity$
$\rho = density$
$\sigma = surface\ tension$
$C_{pl} = specific\ heat$
$C_{sf} = experimental\ constant$
$Pr_l = Prandtl\ number$
$n = experimenatl\ constant$

Putting whole variables of our test conditions into the above equation, especially 1 ℃ on T_{sur}-T_{sat}, then we can get the heat flux for sub-cooled boiling in the test environment, $1.10 \cdot 10^4$ W/m^2.

Finally, we can gather these results into a single plot by inverse operation from CHF calculation to nucleate boiling heat flux calculation to get the temperature difference when the heat flux reaches CHF. The result of abovementioned process, is plotted as below Fig. 1.

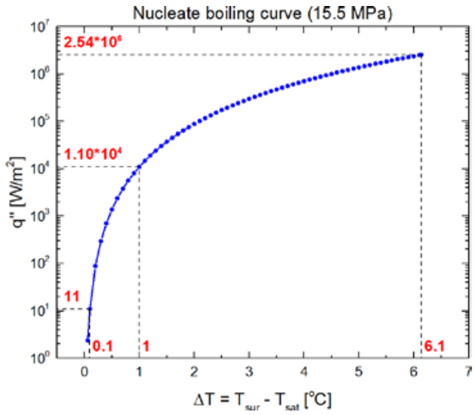

Fig. 1. Nucleate boiling curve for water in 15.5 MPa condition

According to the above graph, it can be said that our setting for sample surface temperature is safe and proper, since it has an engineering margin from the minimum temperature difference and it is not close to the maximum temperature difference.

2.3 Heater Transfer Coefficient

At last, we must calculate heater power to realize our plan. As inlet and outlet temperature of autoclave was set under considering ratio of the test sample dimension and the real fuel cladding dimension, the total power needed to heat the coolant can be calculated like below equation 3.

$$\dot{Q} = \dot{m}C_p(T_{out} - T_{in}) \quad (2)$$

This total power is caused by flow boiling which is combined phenomenon of forced convection and pool boiling. In the previous section, we already get the heat flux of nucleate boiling of pool boiling situation. Therefore, finding the heat flux for forced convection is just left.

The heat flux for forced convection can be calculated by Newton's cooling law, equation 4 below. As the equation shows, we need to find heat transfer coefficient only to calculate heat flux for forced convection.

$$\dot{q} = h \cdot (T_s - T_\infty) \quad (3)$$

Heat transfer coefficient can be calculated from Nusselt dimensionless number, below equation 5.

$$Nu = \frac{D_H h}{k} \quad (4)$$

$D_H = hydraulic\ diameter$
$k = thermal\ conductivity$

There are several ways to find Nusselt number, but the model of Gnielinski et al. in 1976 is known as the most reliable empirical equation, below equation 6 [4]. Thus, in this study, we chose Gnielinski's model to calculate Nusselt number and further to get the heat flux for forced convection on the heated surface.

$$Nu = \frac{\left(\frac{f}{8}\right)\cdot(Re-1000)\cdot Pr}{1+12.7\left(\frac{f}{8}\right)^{\frac{1}{2}}\cdot\left(Pr^{\frac{2}{3}}-1\right)} \quad (5)$$

$f = friction\ factor$
$Re = Reynolds\ number$

3. Experimental

CRUD adhesion test was conducted under the typical water chemistry condition inside of PWR primary circuit as listed below.

Table. 1. Experimental conditions

	PWR	Test Condition
Pressure	15.5 MPa	15.5 MPa
Coolant Temperature	320 ℃	323.5 ℃
Boron	1200 ppm	1200 ppm
Lithium	2 ppm	2 ppm
Dissolved Hydrogen	25 cc/kg	25 cc/kg
Dissolved Oxygen	< 10 ppb	< 10 ppb
pH	6.8 – 7.4	6.8 – 7.4

To generate sub-cooled boiling phenomenon on the heated surface, rod type heater was used. The target surface temperature of rod heater was set as 346 ℃, calculated in previous chapter, which fits with sub-cooled boiling condition in 15.5 MPa and also satisfies IAEA regulation for cladding surface temperature. The material type of rod heater was SS316. The dimensions and power related specification is listed below

Table. 2. Rod heater specification

Material	SS316
Heated Length	300 mm
Total Power	1 kW
Heat Flux	0.167 MW/m^2

There are two major conditions to make CRUD on cladding surface. The first one is sub-cooled boiling phenomenon which is already mentioned and explained. And second one is corrosion product, such as Nickel and Iron. In the real environment, these corrosion products are supplied from steam generator, however, in the test condition, these should be injected directly to test loop equipment.

In this experiment, we referred EPRI technical report to set our metal ion injecting concentration. As the results of Westinghouse advanced loop tester (WALT), average CRUD thickness and porosity were calculated, and also composition of CRUD was measured. We supposed this amount of CRUD will be formed in our case and got how much metal ion concentration are needed to make CRUD on the rod heater surface inversely. The final value we used in this test was 24.82 ppm for Nickel, and 11.75 ppm for Iron respectively [5].

The test had been conducted during 8 days measuring ion conductivity every day by using inductively coupled plasma (ICP). CRUD figures were captured by optical microscope (OM), and scanning electron microscope (SEM) respectively. CRUD composition was measured by energy-dispersive X-ray spectroscopy (EDS).

4. Result & Discussion

After passivating whole loop system, the experiment was started. So, decreasing metal ion concentration could be interpreted as deposition on the heated surface, in this case rod heater surface. According to the result of ICP measurement, Nickel and Iron ion concentration had decreased as below Fig. 2.

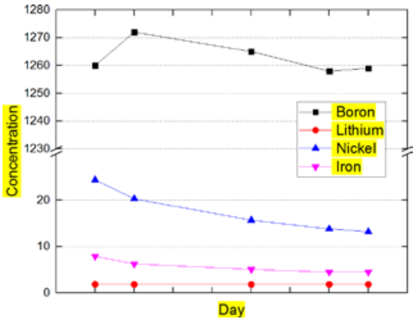

Fig. 2. ICP measurement during 8 days

After cutting sample to observe its surface, OM, and SEM measurements were done. In the case of OM, there was no significant pore on the rod heater surface. From WALT test, pores were observed from the surface. May this difference be caused by testing period.

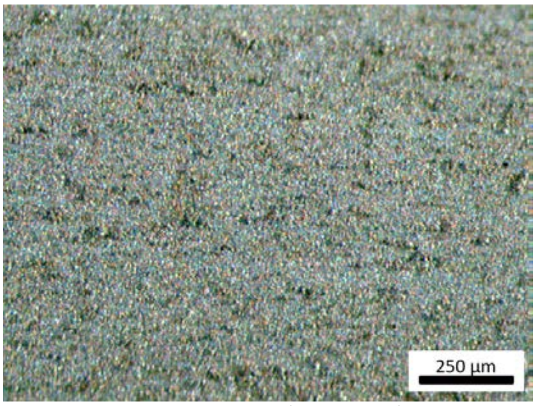

Fig. 3. OM observation result of heater surface

To figure out CRUD formation more precisely than OM result, SEM observation was done. From roughly captured image in 2500 X magnification, there were something strange on the typical Iron oxide layer whose color is white.

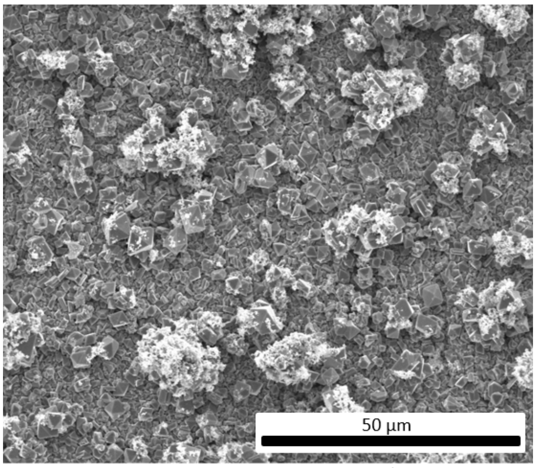

Fig. 4. SEM observation in 2500 X magnification

We thought that white one is some deposition on the heater surface, since this seems like CRUD particles from WALT test cases. So we focused more on it to compare dark gray oxide layer. The magnification increased up to 5000 X to see each layer. According to this try, two different types of oxide were investigated. The one, dark gray colored, seemed like the typical oxide spinel of SS316 material. This oxide layer could be said as passivated oxide layer of rod heater we used in this experiment. The other one, white colored and small size particle, looked like not a normal oxide, since its size is smaller than the previous one, and it was deposited on the SS316 oxide layer as below figure showed us.

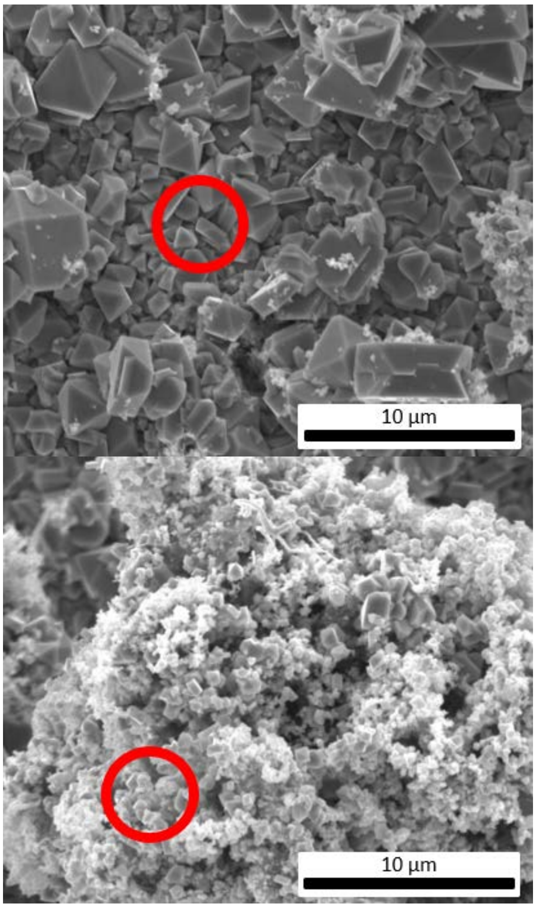

Fig. 5. SEM observation in 5000 X magnification

The red circled region was investigated by EDS to measure the composition of each oxide layer. This could be an exact evidence whether the white one is deposition or not, however, EDS results were not precise. This problem may be lied on the depth gap between typical oxide layer and deposited layer on it. Since EDS resolution is not that much precise.

Therefore, to compare both, other equipment which has higher resolution than SEM/EDS should be needed such as transmission electron microscope (TEM).

Table. 3. EDS result in two different regions

[at %.]	Oxide	Deposit
O	16.14	38.61
Mn	1.65	-
Cr	7.48	1.43
Ni	18.70	19.63
Fe	54.53	40.06
Co	1.49	-
Sn	-	0.28

5. Conclusions

Several heat transfer and thermohydraulic calculations were conducted to set the test conditions for CRUD adhesion test which can simulate real environment of primary circuit water chemistry of commercial nuclear power plant. And CRUD adhesion test was conducted in laboratory scale which can simulate CRUD deposition on the cladding surface.

The heat flux to generate sub-cooled boiling and to heat the coolant to the target temperature was calculated by using various equations. To get heat flux of flow boiling, the summation of forced convection and pool boiling phenomenon was done in point of heat flux estimation.

CRUD adhesion test was conducted during 8 days under the typical water chemistry environment of PWR primary circuit. To track the change of ion concentration we injected to supply corrosion product to loop system, ICP measurements were done which is the evidence of deposition in heated surface.

CRUD figure was observed by OM and SEM respectively, but in case of OM, there was no significant pore structure on the surface. This may be caused by short period of experiment.

SEM images showed some deposited particles on the passivated oxide layer which could be interpreted as CRUD, however, EDS results could not back up this SEM observation because of its low resolution.

In the near future, to figure out whether this kind of deposition is CRUD or not, the experiment will be done more than 8 days. Referring previous researches, 2 weeks seem adequate. And TEM observation will be conducted parallel with SEM/EDS observation to make sure the thickness of CRUD and also its composition.

ACKNOWLDEGEMENT

This work was financially supported by the International Collaborative Energy Technology R&D Program (No. 20168540000030) of the Korea Institute of Energy Technology Evaluation and Planning (KETEP) which is funded by the Ministry of Trade Industry and Energy and was supported by “Human Resources Program in Energy Technology” of the Korea Institute of Energy Technology Evaluation and Planning (KETEP), granted financial resource from the Ministry of Trade, Industry & Energy, Republic of Korea (No. 20174030201430).

REFERENCES

[1] N. E. Todreas, and M. S. Kazimi, Nuclear Systems Thermal Hydraulic Fundamentals, CRC Press, 2012

[2] J. Peybernes, Evaluation of the Forces Generated by Cross-flow on PWR Fuel Assembly, Proceedings of a Technical Meeting Held in Cadarache, France, IAEA-TECDOC-1454, pp.13-22, Nov.22-26, 2004

[3] W. M. Rohsenow, A Method of Correlating Heat Transfer Data for Surface Boiling of Liquids, ASME Transactions 74, pp.969975, 1952

[4] V. Gnielinski, New Equations for Heat and Mass Transfer in Turbulent Pipe and Channel Flow, International Chemical Engineering 16, pp.359-368, 1976

[5] J. Deshon, Simulated Fuel Crud Thermal Conductivity Measurements Under Pressurized Water Reactor Conditions, EPRI Technical Report, 1022896, 2011