Experimental Investigation of the Fouling Characteristics of the CRUD on Pressurized Water Reactor Operating Conditions

Ji Yong Kim, Yunju Lee, Byeongju Kim, Ji Hyun Kim, and In Cheol Bang* Department of Nuclear Engineering, Ulsan National Institute of Science and Technology, 50 UNIST-gil, Ulju-gun, Ulsan, 44919, Republic of Korea *Corresponding author: icbang@unist.ac.kr

Keywords: CRUD (Chalk river unidentified deposit), Sub-cooled nucleate boiling, PWR (Pressurized water reactor), Thermal resistance, Fouling

1. Introduction

The Chalk River Unidentified Deposit (CRUD) has been pointed out as one of the operating issues from the viewpoint of the operational safety and economics of the nuclear power plant [1-3]. Various experimental [4-7] and computational [8-11] research activities were conducted to analyze and predict the CRUD growth behavior and its implications on the fuel cladding heat transfer capability. However, the experimental database related to the CRUD, which was conducted under the pressurized water reactor (PWR) operating condition, available in the open literature for model development and validation is limited to the WALT (Westinghouse Advanced Loop Test) facility due to the difficulty of experiment in high pressure and temperature operating conditions. In order to support the various research and development activities and establish the regulatory basis for the CRUD in operational PWR, mutual verification database expansion of the CRUD-related and experimental results of the existing WALT facility are required. In this study, we present the experimental analyzed CRUD deposition characteristics and fouling resistance behavior in PWR operating conditions using the experimental facility called DISNY crud Deposition Simulator for Nuclear energY).

2. Experimental Setup

2.1 Experimental Facility

The DISNY which is designed and constructed to simulate the prototypical pressurized water reactor operating conditions were utilized in the current experimental study [12]. The operating pressure of DISNY was controlled by a pressurizer and the test section inlet temperature was modulated by preheater operation. The design features of the DISNY facility can be found in Fig. 1.

The Joule-heated heater assembly with an active heating length of 300mm was used in the experiment. The active heating length region of the heater assembly was made of Zr-Nb-Sn alloy cladding with an outer diameter of 9.5mm and an inner diameter of 8.3mm which is the same as the actual fuel cladding for the nuclear reactor to reduce the distortion of the physicochemical reactions related to the CRUD growth.

Two k-type thermocouples and voltage measurement lines are embedded inside the active heating element to measure the heater's inner wall temperature and voltage difference across the active heating length respectively. The schematics of the heater assembly can be found in Fig. 2.



Fig. 1. Design features of the DISNY facility [12].



Fig. 2. Schematics of the DISNY heater assembly [12].

2.2 Experimental Procedure

The experiment was conducted in three steps. At first, we analyze the sub-cooled nucleate boiling (SNB) heat transfer characteristics of the bare cladding surface which are not covered with simulated CRUD. And then, the CRUD growth simulation was conducted under the designated water chemistry conditions within the assigned deposition period. Finally, we analyze the SNB characteristics of the CRUD-deposited cladding surfaces under the PWR operating conditions. The experiment was conducted in an in-situ way to minimize the delamination or degradation of the simulated CRUD during the experiment. The borated water with 1,200 ppm of boron and 2.2 ppm of lithium was used as a working fluid in the experiment. The thermal-hydraulic and water chemistry conditions of the current experimental study to simulate the CRUD growth under operating PWR are summarized in Table I. The detailed reasonings and backgrounds related to the

selection of the experimental conditions can be found in our group's previous work [12].

Thermal-hydraulic conditions			
Parameters	P _{PZR}	T_{in}	Mass flux
Values	155.17	336.82	3,435.18
	[bar]	[°C]	$[kg/m^2-s]$
Water chemistry conditions			
Parameters	Ion	Deposition	Deposition
	concentration	heat flux	time
Values	Ni: 36 [ppm]	477.22	21.73
	Fe: 18 [ppm]	$[kW/m^2]$	[hr]

Table I: Summary of the experimental conditions.

2.3 CRUD Growth Simulation Methodology

The Fe-EDTA (Ethylene-Diamine-Tetra acetic Acid) and Ni-EDTA were used as CRUD growth precursors to simulate the corrosion products in actual PWR. The upper mentioned chemicals are injected into the DISNY loop by pressurizing pump during the CRUD deposition simulation period. The injected CRUD precursors are expected to be decomposed into Fe and Ni ions form in high-pressure and temperature working conditions [13] of the DISNY facility and forms particulate corrosion products such as NiFe₂O₄, Fe₂O₃, and NiO. The target concentration of the metal ion precursors was increased significantly compared to the PWR normal operating water chemistry condition to reduce the CRUD deposition time frame within the week.

2.4 Data Reduction Methodology

The SNB curves before and after the CRUD deposition and fouling resistance during the CRUD growth simulation were evaluated in the current experimental study. The upper-mentioned quantities were selected as key parameters to investigate the effects of CRUD deposition on the SNB characteristics of the zirconium-based alloy cladding under the PWR operating conditions. The applied heat flux was evaluated by Ohm's law as depicted in Eqn. (1). Where q'' is the applied heat flux, ΔV_{clad} is the voltage drop across the active heating length of the heater, I is the applied electrical current, and A_{clad} is the heat transfer area of the active heat ling length. The SNB heat transfer coefficient (h) of the zirconium-based alloy cladding was evaluated followed by Eqn. (2). Where, T_{o} is outer cladding temperature, which is calculated based on Furrier's Law of conduction with consideration of the volumetric heat source q''' in the cylindrical coordinate system, and T_{sat} is the saturation temperature of working fluid at a given operating pressure condition. The fouling resistance (R_f) of the CRUD deposited cladding surfaces are evaluated based on the Eqn. (4). Where, h_0 is the heat transfer coefficient value at the initial stage of CRUD deposition simulation (without

CRUD), and h(t) is the time-dependent heat transfer coefficient value during the CRUD deposition simulation period.

$$q'' = \frac{\Delta V_{clad} I}{A_{clad}} \qquad \text{Eqn. (1)}$$

$$h = \frac{q''}{T_o - T_{sat}} \qquad \qquad \text{Eqn. (2)}$$

$$T_{o} = T_{i} + \frac{q'''}{4k_{clad}} \left(r_{i}^{2} - r_{o}^{2}\right) - \frac{q'''}{2k_{clad}} r_{i}^{2} \ln\left(r_{i} / r_{o}\right) \text{ Eqn. (3)}$$

$$R_f = \frac{1}{h_o} - \frac{1}{h(t)} \qquad \text{Eqn. (4)}$$

The uncertainty analysis of the evaluated parameters such as applied heat flux, cladding outer wall temperature, heat transfer coefficient, and fouling resistance were evaluated based on the error propagation method [14] as depicted in Eqn. (5). Where U is the uncertainty of the given parameters, δ derived variables, x is the parameter that affects the given variable. The calculated uncertainty for the heat flux, cladding outer temperature, heat transfer coefficient, and fouling resistance are given as $\pm 0.58\%$, $\pm 1.62\%$, $\pm 1.72\%$, and $\pm 3.44\%$ respectively.

$$\frac{U_{\delta}}{\delta} = \frac{1}{\delta} \sqrt{\sum_{i=1}^{n} \left(\frac{\partial \delta}{\partial x}\right)^{2} U_{x}^{2}} \qquad \text{Eqn. (5)}$$

3. Results and Discussions

3.1 Sub-cooled Nucleate Boiling without CRUD

The averaged SNB curve of the DISNY heater before the CRUD deposition simulation (Bare surface; without CRUD layer) is shown in Fig. 3. At the applied heat flux condition lower than ~300kw/m², the cladding superheat increases linearly with increasing the heat flux which indicates that the heat removal mechanism of the active heating length is governed by the forced convection. Around the applied heat flux of ~300kW/m², the slope of the boiling curve increases rapidly which means that the efficiency of the heat transfer increase significantly and indicates the onset of nucleate boiling (ONB).

The comparison between the boiling curve trends from the DISNY facility and the WALT facility [7], which is the representative established one to analyze the thermal resistance characteristics of the CRUD, was conducted to validate the performance of the DISNY facility to simulate SNB phenomena under PWR operating conditions. As shown in Fig. 3, the boiling curve data from both DISNY and WALT facilities shows similar tendencies under the same heat flux conditions, while a slight difference in wall superheats exists. However, the degree of the difference in wall superheat in the boiling curves is small and can be neglected.

The performance of the DISNY facility was validated based on the comparison results of the boiling curve data from both the DINSY and WALT facilities. Therefore, a further experimental procedure proceeded to simulate the CRUD growth and to evaluate the thermal resistance characteristics of the simulated PWR CRUD.



Fig. 3. Sub-cooled nucleate boiling curves from DISNY facility and comparison results between WALT facility [7].

3.2 Sub-cooled Nucleate Boiling with CRUD

The photo of the retrieved heater specimen from the DISNY facility after the experiment is presented in Fig. 4. As shown in Fig. 4, the dark brown layer, which is simulated CRUD formed during the current experiment, can be found on the zirconium-based alloy cladding.



Fig. 4. Photo of retrieved heater specimen after the experiment.

The boiling curve data for bare cladding surfaces and CRUD-deposited cladding surfaces are shown in Fig. 5. The boiling curves for the bare cladding surface without the CRUD layer correspond to black and red lines, The boiling curves for the CRUD-deposited claddings are depicted in blue and green line in the boiling curve. One of the distinguished features of the CRUDdeposited zirconium-based alloy cladding surface's boiling curve is the ONB difference. For the CRUD- deposited cladding case, the heat flux value corresponding to the ONB is much lower than the bare case. The difference in the ONB is attributed to the fact that the simulated CRUD layer under the single-phase convection condition acts as an additional thermal resistance and increases the outer cladding wall temperature of the cladding as depicted in the low heat flux region of the boiling curve. Due to the increased thermal resistance, the CRUD-deposited surface shows higher wall superheat under the same applied heat flux condition compared to the bare surface. Therefore, the required wall superheat for the bubble formation (ONB) can be met at relatively lower heat flux conditions for the CRUD-deposited cladding case. For the applied heat flux range of ~250–450kW/m², no big differences were observed from the boiling curves of both bare and CRUD-deposited cladding cases. After the heat flux reached \sim 500kW/m² or even larger, the wall superheat of the CRUD-deposited cladding surfaces showed a higher value compared to the bare case. The cladding outer temperature difference between bare and CRUDdeposited cladding surfaces becomes greater along with the increase of the applied heat flux.

In general, the surface temperature of the zirconiumbased alloy cladding with a simulated CRUD layer tended to show higher values than that of the bare cladding surface. Also, it was experimentally confirmed that the effect of fouling thermal resistance on the fuel cladding heat transfer is affected by the applied heat flux to the cladding and the governing heat removal mechanism such as single-phase convection or SNB.



Fig. 5. Boiling curve difference between bare and CRUD deposited zirconium-based alloy cladding surfaces.

3.3 Fouling Resistance Characteristics of CRUD

Fig. 6 shows the cladding surface temperature behavior and calculated fouling resistance value of the zirconium-based alloy cladding during the CRUD growth simulation period of the experiment. The cladding outer surface temperature and the calculated fouling resistance decreased until 10 hours after the start of the CRUD deposition simulation, and after the 10 hours of deposition, it showed a tendency to recover. The behavior of decreasing wall superheat and fouling resistance at the beginning of the CRUD deposition simulation is related to the increased surface roughness as micron or sub-micron scale corrosion products are deposited on the surface of the cladding in very thin thickness and form an interlinked primitive porous fouling layer [15,16]. As the CRUD deposit thickness increase during the growth simulation period, the thermal resistance effect of CRUD become dominant rather than the local heat transfer enhancement due to the increased surface roughens, the cladding surface temperature, and the calculated fouling resistance increase again. The microstructure analysis and the thickness measurement analysis, which will be planned to be conducted, for the simulated CRUD are required to strongly support the upper mentioned reasonings.



Fig. 6. Calculated fouling resistance of fuel cladding surface during CRUD deposition simulation.

4. Summary and Conclusion

In this study, the thermal resistance characteristics of the simulated CRUD under the pressurized water reactor operating conditions were experimentally investigated with the DISNY facility. The performance of the DISNY facility was validated by comparing the sub-cooled nucleate boiling curves with the WALT facility. After performance validation, the CRUD growth simulation was conducted to investigate the subcooled nucleate boiling characteristics of the CRUD deposited cladding surface. The deposition of the CRUD on the fuel cladding surfaces increases the wall superheat during both single and two-phase flow conditions by acting as an additional thermal barrier to the heat transfer process of the cladding. The fouling resistance characteristics of the simulated CRUD during the CRUD growth period were analyzed.

The experimental analysis results of simulated CRUD under the pressurized water reactor operating conditions show that the CRUD can act as additional thermal resistance to the cladding heat transfer and increase the temperature level on both single and twophase flow conditions. Increased operating temperature of fuel cladding by CRUD deposition can accelerate the corrosion behavior and in some severe cases can cause the failure of nuclear fuel by CRUD-induced localized corrosion mechanism [3,17]. The research activities from the DISNY facility are expected to give physical insight into CRUD-related safety issues and can be acts as a valuable database for the development and validation of computational models and regulatory guidelines.

ACKNOWLEDGEMENT

This work was supported by the Nuclear Safety Research Program through the Korea Foundation Of Nuclear Safety (KoFONS) using the financial resource granted by the Nuclear Safety and Security Commission (NSSC) of the Republic of Korea. (No. 2106022)

REFERENCES

[1] P. Saha, N. Aksan, J. Andersen, J. Yan, J. P. Simoneau, L. Leung, F. Bertrand, K. Aoto, H. Kamide, Issues and future direction of thermal-hydraulics research and development in nuclear power reactors, Nuclear Engineering and Design, Vol.264, p. 3–23, 2013.

[2] K. Shirvan, Implications of accident tolerant fuels on thermal-hydraulic research, Nuclear Engineering and Design, Vol.358. 110432, 2020.

[3] J. Deshon, D. Hussey, B. Kendrick, J. Mcgurk, J. Secker, M. Short, Pressurized Water Reactor Fuel Crud and Corrosion Modeling, JOM., Vol.63, pp. 64–72, 2011.

[4] S. H. Baek, H. S. Shim, J. G. Kim, D. H. Hur, Effects of heat flux on fuel crud deposition and sub-cooled nucleate boiling in simulated PWR primary water at 13 MPa, Annals of Nuclear Energy, Vol.133, pp.178–185, 2019.

[5] J. Ham, Y. Lee, S. C. Yoo, M. P. Short, C. B. Bahn, J. H. Kim, Effect of TiN Coating on the Fouling Behavior of Crud on Pressurized Water Reactor Fuel Cladding, Journal of Nuclear Materials, Vol.549, 152870, 2021.

[6] I. Dumnernchanvanit, N. Q. Zhang, S. Robertson, A. Delmore, M. B. Carlson, D. Hussey, M. P. Short, Initial experimental evaluation of crud-resistant materials for light water reactors, Journal of Nuclear Materials, Vol.498, pp.1-8, 2018.

[7] G. Wang, A. Byers, M. Young, Simulated Fuel Crud Thermal Conductivity Measurements Under Pressurized Water Reactor Conditions, EPRI, Palo Alto, CA, 2011.

[8] D. Y. Yeo, H. C. NO, Modeling heat transfer through chimney-structured porous deposit formed in pressurized water reactors, International Journal of Heat and Mass Transfer, Vol.108, pp.868–879, 2017.

[9] M. P. Short, D. Hussey, B.K. Kendrick, T. M. Besmann, C. R. Stanek, S. Yip, Multiphysics modeling of porous CRUD deposits in nuclear reactors, Journal of Nuclear Materials, Vol.443(1-3), pp.579-587, 2013.

[10] C. Pan, B. G. Jones, A. J. Machiels, Concentration levels of solutes in porous deposits with chimneys under wick boiling conditions, Nuclear Engineering and Design, Vol.99(1), pp.317-327, 1987.

[11] P. Cohen, HEAT AND MASS TRANSFER FOR BOILING IN POROUS DEPOSITS WITH CHIMENYS, AlChe Symphsium, Ser. 70, pp. 71–80, 1972.

[12] J. Y. Kim, Y. Lee, J. H. Kim, I. C. Bang, The DISNY facility for sub-cooled flow boiling performance analysis of CRUD deposited zirconium alloy cladding under pressurized water reactor condition: Design, construction, and operation, Nuclear Engineering and Technology, Vol.55(9), pp. 3164-3182, 2023.

[13] W. A. Byers, G. Wang, M. Y. Young, J. Deshon, SIMULATION OF PWR CRUD, Proceedings of 22nd International Conference on Nuclear Engineering (ICONE22), Prague, Czech Republic, 2014.

[14] H.W. Coleman, W.G. Steele, Experimentation, validation, and uncertainty analysis for engineers, John Wiley & Sons, 2018.

[15] S. M. Peyghambarzadeh, A. Vatani, M. Jamialahmadi, Application of asymptotic model for the prediction of fouling rate of calcium sulfate under subcooled flow boiling, Applied Thermal Engineering, Vol.39, pp.105-113, 2012.

[16] O. P. Arsenyeva, B. Crittenden, M. Yang, P. O. Kapustenko, Accounting for the thermal resistance of cooling water fouling in plate heat exchangers, Applied Thermal Engineering, Vol.61(1), pp.53-59, 2013.

Engineering, Vol.61(1), pp.53-59, 2013. [17] D. Y. Yeo, A. J. Huxford, V. Petrov, A. Manera, W. Gurecky, B. S. Collins, DEVELOPMENT OF A TWO-PHASE FLOW MODEL IN MAMBA FOR CRUD-INDUCED LOCALIZED CORROSION MODELING CAPABILITY, Proceedings of 19th International Topical Meeting on Nuclear Reactor Thermal Hydraulics (NURETH-19), Brussels, Belgium, 2022.