

Evaluation of the Corrosion Behavior of the Aluminum Cladding in the KMRR Fuel

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KMRR 핵연료 알루미늄 피복재의 부식 거동 평가

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Abstract

For the evaluation of the corrosion behavior of the aluminum cladding in the KMRR(Korea Multi-purpose Research Reactor) fuel, a modified Griess correlation was derived by introducing a heat flux factor derived from the comparison of the measured in-reactor corrosion data with the prediction of the Griess correlation. As a design criterion on the corrosion to maintain the KMRR fuel integrity, prevention of the oxide spallation was conservatively selected, which is conservatively assumed to occur when the temperature difference across the oxide layer exceeds 114°C . A bounding power history of the KMRR fuel was determined by examining all the power histories of the KMRR fuel from cycle 1 to equilibrium cycle, and used to predict the maximum possible corrosion. Results of the corrosion prediction of the KMRR fuel with the bounding power history showed that the maximum local thickness of the oxide layer would be below $50\mu\text{m}$ and the design criterion on the oxide spallation would be satisfied with a factor of two margin. Therefore, it can be said that corrosion of the cladding will not impair the integrity of the KMRR fuel. Nevertheless, the applicability of the modified Griess correlation to the KMRR needs to be further verified through the KMRR fuel corrosion surveillance.

요 약

KMRR(다목적 연구용원자로) 핵연료의 알루미늄 피복재의 부식거동을 평가하기 위해, 부식 예측치와 노내 부식 실측치의 비교를 통해 유도된 열속인자를 도입한 수정된 Griess 경험식을 유도하였다. KMRR 핵연료의 건전성이 유지되는 부식의 설계기준으로써 산화층의 박리 방지가 보수적으로 설정되었으며, 산화층의 박리는 산화층에서의 온도차이가 114°C 이상에서 일어난다고 보수적으로 가정하였다. KMRR 핵연료의 출력이력을 첫 주기부터 평형주기까지 분석하여, 한계출력이력을 결정하였다. 한계출력이력을 가진 KMRR 핵연료의 부식량 예측계산 결과, 최대 산화층의 두께는 $50\mu\text{m}$ 이하였으며, 산화층 박리의 설계기준은 2배의 여유도를 가지고 만족하였다. 따라서, KMRR 핵연료는 피복재의 부식

으로 인해 손상되지 않을 것으로 판단된다. 그러나, 수정된 Griess 부식경험식의 KMRR에의 적용 타당성은 KMRR 핵연료의 부식 감시를 통해 추가로 검증될 필요성이 있다.

1. Introduction

Aluminum alloys have been widely used as the cladding material of the research reactor fuels for good thermal conductivity, high ductility and low neutron absorption characteristics as well as high corrosion resistance in the high purity water. As shown in Table 1, several kinds of the aluminum alloys are commercially available for the fuel cladding material. For the KMRR fuel [1], an aluminum alloy, AA-1060 is used as a cladding material, which has been used for more than thirty years as the cladding material in NRU reactor [2].

Corrosion rate of the aluminum alloy cladding is influenced by such variables as the temperatures of the cladding and the coolant, coolant chemistry (pH, additives and impurities, etc.), surface heat flux and corrosion layer thickness. Effect of the corrosion of the aluminum alloy cladding upon the fuel performance are the temperature increase of the fuel and the cladding due to the very low thermal conductivity of the aluminum oxide, which can lead to the loss of the creep resistance of the cladding and the thermal expansion of the fuel, and the fuel failure by the perforation of the cladding due to the excessive corrosion. As the corrosion layer of the aluminum cladding increases above a certain limit, the spallation of the corrosion layer could occur, which may be followed by the severe localized attack in the form of a

subsurface void. As a design limit of the cladding oxidation to maintain the integrity of the KMRR fuel, oxide spallation was conservatively selected even though it does not directly results in fuel failure.

In this study, the corrosion behavior of the aluminum cladding of the KMRR fuel will be evaluated. Test data and correlations on the aluminum cladding corrosion will be reviewed to find an appropriate corrosion prediction method for the KMRR fuel. Then, the bounding power history of the KMRR fuel will be determined to predict the maximum possible corrosion layer thickness to find whether the spallation of the oxide layer would occur.

2. Aluminum Alloy Corrosion

2.1. Literature Survey

A. Griess Correlation

Griess correlation [3, 4, 5, 6] has been developed from the data of the aluminum alloy corrosion tests performed in an out-of-pile test loop of the Oak Ridge National Laboratory (ORNL) to evaluate the corrosion of the aluminum cladding in the High Flux Isotope Reactor (HFIR) and the Advanced Test Reactor (ATR) conditions [3, 4, 5, 6]. The test conditions of the experiments are shown in Table 2. Majority of the tests were performed at the pH 5.0

Table 1. Chemical Composition of Aluminum Alloys

Alloy	Element (wt. %)						
	Ni	Fe	Mg	Si	Cu	Cr	Zn
AA-1100		0.45		0.04	0.11		
AA-6061		0.41	1.15	0.39	0.24	0.18	0.03
X-8001	1.14	0.55		0.03			
AA-1060		0.35	0.03	0.25	0.05		0.05

which is the operation condition of the HFIR and the ATR, and only several tests were performed at the pH range of 5.7-7.0. Three kinds of the aluminum alloys such as AA-1100, X-8001 and AA-6061 were tested, and it was found that there was no significant difference in the corrosion behavior among them. Tests at the pH 5.0 showed less corrosion than those at the pH 5.7-7.0. The test results at the pH 5.0 showed the dependence on the cladding surface temperature and the time, and the test results at the pH 5.7-7.0 showed the same dependence. From the analysis of the test results, Griess derived the following correlation.

$$X_G(t) = 11252 t^{0.778} \exp(-4600/K), \quad \text{pH 5.0} \\ 30480 t^{0.778} \exp(-4600/K), \quad \text{pH 5.7-7.0}$$

where,

$$X_G(t) = \text{corrosion layer thickness } (\mu\text{m}) \\ t = \text{time (hour)} \\ K = \text{cladding surface temperature (K)}.$$

Griess correlation could be best used in the range of the heat flux between 3.15 MW/m² and 6.3 MW/m²

[6]. It was also found [6] that at the heat flux below 3.15 MW/m², significantly lower oxidation of the aluminum alloy than the prediction of the Griess correlation was measured such that the corrosion rate at the heat flux of 1.6 MW/m² was about half of that at the heat flux of 3.15-6.3 MW/m². It needs to be noted that a majority of the corrosion tests were performed above the heat flux of 4.7 MW/m² which was considered to be the peak heat flux of the ATR. [6]

B. ANSR Correlation

ANSR correlation [7, 8] was developed from the results of the AA-6061 aluminum alloy corrosion tests performed in the out-of-pile test loop in ORNL to evaluate the corrosion of the fuel cladding in the Advanced Neutron Source Reactor (ANSR) [7]. As shown in Table 2, the tests were performed at the condition of high temperature and high heat flux which was comparable to the operation condition of the ANSR. ANSR correlation which predicts the corrosion of the AA-6061 aluminum alloy as a function of the temperature, heat flux and the time is as

Table 2. Comparison of Test Conditions which the Corrosion Correlations of the Aluminum Alloys are Based Upon with the KMRR Operation Condition

correlation	pH	coolant temperature (°C)	surface temperature (°C)	coolant velocity (m/sec)	heat flux (MW/m ²)	duration (days)	place	aluminum alloy	reference reactor
Griess correlation	5.0 and 5.7-7.0	T _{local} = 54-121	121-204	7.6-15.2	3.2-6.3	10-17	out-of-pile loop	AA-6061 AA-1100 X-8001	HFIR ATR
ANSR correlation	5.0	T _{in} = 39-49 T _{out} = 44-99	96-201	25-28	6.2-20.2	2-24	out-of-pile loop	AA-6061	ANSR
Hanson correlation	5.0	N/A	79.4-121	13.7	4-5	9.2-47.9	ATR operation data	AA-6061	ATR
Kritz correlation	5.0	N/A	N/A	N/A	2.2(max.)	N/A	SRP operation data	AA-6061	SRP
KMRR operation condition	5.5-6.5	T _{in} = 35 T _{out} = 45	50-125	7.2	1.27(avg.) 3.15(max.)	180-210	KMRR operation	AA-1060	KMRR

follows.

$$X_A(t) = [X_{A0}^{n+1} + (n+1)kt]^{1/(n+1)}$$

where,

- $X_A(t)$ = corrosion layer thickness (μm)
- X_{A0} = initial corrosion layer thickness (μm)
- t = time (hours)
- k = rate constant ($\mu\text{m}^{n+1}/\text{hour}$)
- n = constant

The rate constant, k defined as a function of the local coolant temperature and the heat flux is

$$k = 6.992 \times 10^5 \exp[-7592/(T_c + 10\Phi)] \mu\text{m}^{1.351}/\text{hour}$$

where,

- T_c = local (bulk) coolant temperature (K)
- Φ = heat flux (MW/m^2)

And a time exponent, $1/(n+1)$ is derived as 0.74 ± 0.07 from the test results. Then, when the initial corrosion layer thickness is zero, a nominal ANSR correlation becomes

$$X_A(t) = 2.64 \times 10^4 t^{0.74} \exp[-5618/(T_c + 10\Phi)],$$

where,

$$X_A(t) = \text{corrosion layer thickness } (\mu\text{m}).$$

C. Hanson Correlation

Hanson correlation [9] was developed through the statistical analysis of the measurement results of the corrosion of the AA-6061 aluminum alloy cladding irradiated in the Advanced Test Reactor (ATR). Nineteen measured data points were obtained from the high burnup fuels of which the cladding temperatures were about 250°F (121.1°C) and 175°F (79.4°C) during the irradiation and the irradiation time ranges from 220 hours to 1150 hours. Hanson correlation which predicts the corrosion of the alumi-

num alloys as a function of the cladding surface temperature and the time is as follows.

$$X_H(t) = 60.782 t^{0.2578} \exp(-2412.5/R)$$

where,

- $X_H(t)$ = corrosion layer thickness (μm)
- t = time (hours)
- R = cladding surface temperature (R)

It was reported [9] that the standard deviation was about 31% when the prediction of the Hanson correlation was compared with the measured corrosion data.

D. Kritz Correlation

Kritz correlation [10] was developed for the application to the Savannah River Production Reactor (SRP) and has a similar form to the Griess correlation except for a different activation energy and an additional term correcting variation in heat flux.

$$X_K(t) = 6.531 q'' t^{0.778} \exp(-1880/K)$$

where,

- $X_K(t)$ = corrosion layer thickness (μm)
- q'' = heat flux (MW/m^2)
- t = time (hour)
- K = cladding surface temperature with the water (K)

2.2. Derivation of the Aluminum Alloy Corrosion Correlation for the KMRR Fuel

Since there is no corrosion data available under the specific operation condition of the KMRR, a literature survey and analysis of the aluminum alloy corrosion data available is performed to find an appropriate corrosion prediction method for the KMRR fuel. As explained in section 2.1, all the corrosion correlations of the aluminum alloy, available in a lit-

erature were developed under a specific thermal-hydraulic condition to be applied to the corresponding research reactors. Corrosion of the aluminum alloy is influenced by such variables as cladding temperature, coolant temperature, surface heat flux, coolant chemistry and corrosion layer thickness, etc. Coolant chemistry includes pH, additives and impurities. It was reported [8] that corrosion of the aluminum alloy could be significantly influenced by the elements in the coolant released from the materials in the cooling system. Specifically, elements released from the stainless steel in the cooling system may deposit on the aluminum oxide to form a Fe-rich layer and subsequently lower the corrosion rate of the aluminum alloy.

In Table 2, test conditions upon which the corrosion correlations are based are compared with the operation condition of the KMRR. Griess, Kritz and Hanson correlations have the comparable thermal-hydraulic condition to the KMRR whereas the Griess correlation covers the pH range of the KMRR. The ANSR correlation is based upon the heat flux much higher than the KMRR condition.

To select the appropriate correlation for the KMRR fuel cladding corrosion, the followings need to be considered.

- Aluminum alloy for the KMRR fuel cladding is AA-1060.
- pH of the KMRR coolant is 5.5-6.5.
- Surface heat flux of the KMRR fuel is 1.27 MW/m² at average and 3.15 MW/m² at maximum.

It has been reported [6] that there was no significant difference in the corrosion behavior among AA-6061, AA-1100 and X-8001. Therefore, considering the relative similarity in the chemical composition between AA-1100 and AA-1060, compared with the difference between AA-1100 and X-8001, it can be assumed that the corrosion behavior of AA-1060, the KMRR fuel cladding material will be similar to those of AA-1100, AA-6061 and X-8001.

The coolant pH clearly seems to have a major ef-

fect on the corrosion of the aluminum alloy. It was found by Griess [6] that the corrosion rate at the range of pH 5.7-7.0 was 2.7 times higher than that at pH 5.0.

For the thermal-hydraulic conditions such as heat flux, cladding surface temperature and coolant temperature, it was found that surface heat flux as well as cladding surface temperature could influence the corrosion significantly. In Table 3 [12], corrosion measurement from the irradiated fuels from different research reactors were compared with the prediction of the Griess correlation. The ratio of the measured one to the predicted one ranges from 0.2 to 3.0 depending upon the thermal-hydraulic conditions. It was found that the ratio strongly depended upon the surface heat flux.

Griess correlation has been widely used to predict the corrosion of the aluminum alloy fuel cladding in most of the research reactors since it was developed in 1964. However, the one deficiency of the Griess correlation is that it can not correctly predict the effect of the heat flux upon the corrosion of the aluminum alloy which has been known to be significant.

The Griess correlation prediction and the measured data of the aluminum alloy corrosion are compared in Table 3. It can be seen from Table 3 that Griess correlation over-predicts oxide thickness by a factor of 5 at the heat flux of 2.2 MW/m² and under-predict by a factor of 3 at the heat flux of 17 MW/m², and therefore, the heat flux has a significant effect on the aluminum alloy corrosion.

The ratio versus surface heat flux is shown in Figure 1. It was found that the ratio has a linear relation with surface heat flux such that

$$f(q'') = -0.20836 + 0.18915 q'',$$

where,

$$q'' = \text{heat flux (MW/m}^2\text{)}.$$

For this reason a heat flux factor was introduced to the Griess correlation to expand the range of the heat flux in which the Griess correlation can be ap-

plied. To be conservative, a minimum of the heat flux factor was set at 0.2 which corresponds to the SRP data in Table 3. Then, a modified Griess correlation with a heat flux factor becomes

$$X_G(t) = 11252 f(q'') t^{0.778} \exp(-4600/K), \text{ pH } 5.0$$

$$30480 f(q'') t^{0.778} \exp(-4600/K), \text{ pH } 5.7-7.0$$

where,

$f(q'')$ = a heat flux factor.

A heat flux factor is

$$f(q'') = -0.20836 + 0.18915 q'', q'' > 2.16 \text{ MW/m}^2$$

$$0.2, q'' \leq 2.16 \text{ MW/m}^2$$

where,

q'' = heat flux (MW/m^2).

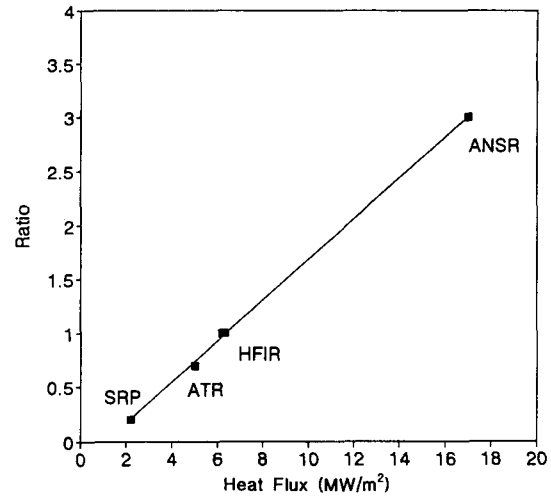


Fig. 1. Ratio of the Measured to the Predicted Oxide Film Thickness by Griess Correlation versus Heat Flux

Table 3. Comparison of Measured Oxide Film Thickness with Thickness Predicted by Griess Correlation [12]

Case No.	Reactor	Years	Coolant	Duration (days)	Velocity(V) (m/sec)	Max. Heat Flux(Φ) (MW/m^2)	Φ/V (MW. sec/m^2)	$\Phi \cdot V$ (MW/m. sec)	$\frac{\delta_{ox}}{(\delta_{ox})_{Griess}}$
1	SRP (production)	76-77	D ₂ O	270	5.8	2.2	0.38	12.8	0.2
2	ATR**	~87	H ₂ O	80-100 (max.)	13.7	< 5; 4 (avg.)	0.29	54.8	0.7
3	HFIR (c. tests)	Early 60s	H ₂ O	20 (max.)	15.2 (max.)	6.3	0.41	95.8	1.0***
4	HFIR (100 MW(t))	Through 11/86	H ₂ O	23	16.2	6.2	0.38	100.4	1.0****
5	ANSR (c. test 4)	88	H ₂ O	5	27.4	12.0	0.44	328.8	2.0~3.0: 2.7?
6	ANSR (design)	88	D ₂ O	14-15	27.4	17.0	0.62	465.8	3?

*: For coolant pH ~5.0.

** : For ATR oxide data base being reevaluated by INEL reactor power < 190 MW(t) : average power ~150 MW(t)

*** : $\pm 25\%$

**** : Presumed; No oxide data obtained shortly after shutdown are available.

2.3. Verification of the Applicability of the Modified Griess Correlation to the KMRR Fuel

To check the validity of the modified Griess correlation in the application to the KMRR fuel, one corrosion measurement datum [13, 14] obtained from the Canadian NRU reactor which uses the same aluminum alloy cladding, AA-1060 and the same coolant pH, 5.5-6.5 as the KMRR, and is operated under similar thermal-hydraulic condition to the KMRR is compared with the prediction of the modified Griess correlation. Measured thickness of the corrosion layer was $14\mu\text{m}$ at the burnup of 36 at. % with the power history shown in Table 4. The prediction by the modified Griess correlation gives $14\mu\text{m}$ while that of the original Griess correlation gives $57.5\mu\text{m}$. There is one more datum from the NRU reactor of which the power history is not known. It showed that at the burnup of 81.4 at. % the measured corrosion layer thickness was $22\mu\text{m}$, which can be assumed to have a similar corrosion rate to the above-mentioned corrosion datum and far less corrosion rate than the Griess correlation predicts. Although the amount of data are inadequate, there was a good agreement between the prediction of the modified Griess correlation and the NRU corrosion data, which can give a limited validation of the modified Griess correlation for the KMRR fuel. To validate the model with statistical significance, it may be still necessary to validate further through the surveillance of the KMRR fuel corrosion.

3. Evaluation of the Corrosion of the Aluminum Alloy Cladding in the KMRR Fuel

3.1. Operation Condition of the KMRR Fuel

KMRR is a pool-type reactor with a forced convection flow of the coolant. KMRR fuel element consists of the Al-61.4% U_3Si fuel meat cladded with the aluminum alloy AA-1060 with 8 circumferentially distributed cooling fins to enhance the fuel element cooling as shown in Figure 2. Two dimensional analysis of the fuel element heat transfer showed that the cooling fins reduce the surface heat flux at least by 25% compared with the case without the cooling fins. There are two kinds of the fuel assemblies, one with 18 fuel elements and the other

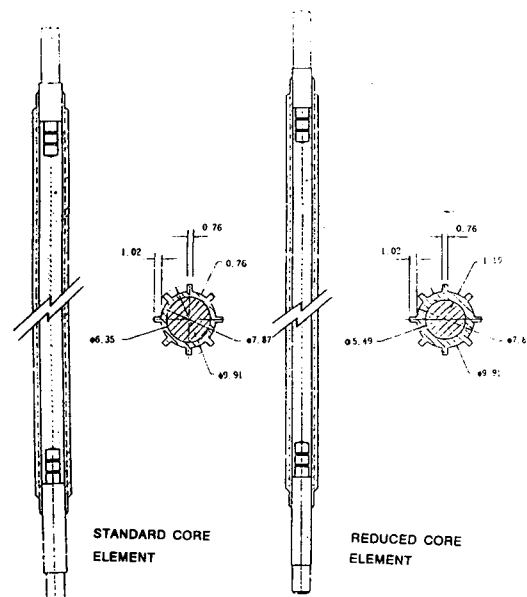


Fig. 2. Schematic of the KMRR Fuel

Table 4. Corrosion Test Result and Power History of the NRU EXP. FZZ-905 [13, 14]

Phase	Time (days)	Power (kW/m)	Heat Flux (MW/m^2)	Fuel Centerline Temperature (K)	Cladding Surface Temperature (K)	Accumulated Burnup (at. %)
I	42	64.5	2.20	421	377	18.4
II	31	75.4	2.57	433	385	36

with 36 fuel elements. Twenty 36-element fuel assemblies and twelve 18-element fuel assemblies are loaded in the core of the KMRR. Each fuel assembly stays for 6 or 7 cycles and the duration of one cycle is thirty days. Thermal-hydraulic conditions of the KMRR core are summarized in Table 5.

3.2. Evaluation of the Corrosion

To predict the corrosion of the KMRR fuel cladding, the power history and the coolant condition have to be determined. To get the maximum conservative corrosion of the fuel cladding, all the power histories of the fuels from cycle 1 to the equilibrium cycle were analyzed and the bounding power history was determined as a function of the burnup by selecting maximum local power among all the power histories at each burnup.

Figure 3 shows a bounding power history and a realistic power history of the KMRR 18-element fuel with the highest burnup. The local power of the KMRR fuel is 104 kW/m at maximum and decreases with the burnup. The 18-element fuel has generally

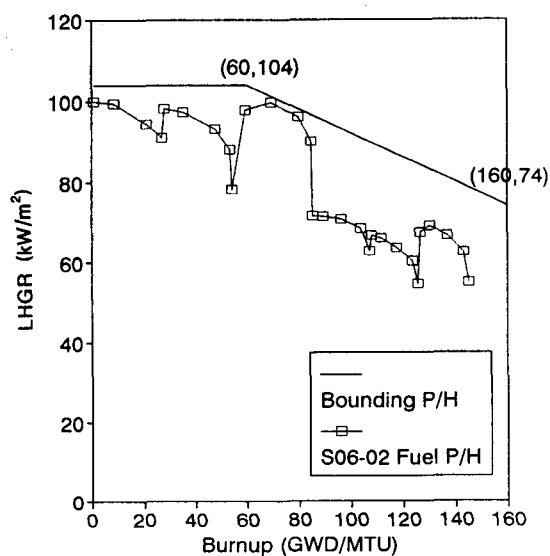


Fig. 3. Bounding and Representative Power Histories of the KMRR Fuel

higher power level than the 36-element fuel. Local coolant temperature and the heat transfer coefficient between the cladding and the coolant was obtained from the sub-channel thermal-hydraulic analysis with a consideration of the axial power distribution.

The modified Griess correlation with the heat flux factor was used to predict the corrosion of the aluminum cladding. Figure 4 shows the corrosion of the KMRR fuel cladding with the bounding and a realistic high burnup power histories. The maximum local thickness of the corrosion layer of the cladding at discharge is less than $50\mu\text{m}$ for the bounding power history and $33\mu\text{m}$ for the realistic high burnup power history. Figure 5 shows the temperature difference over the oxide layer which depends upon the thermal conductivity of the oxide layer and the heat flux. The thermal conductivity of the aluminum oxide layer is $2.25 \pm 0.35 \text{ w/cm}^2$ and the lower limit of 1.9 w/cm^2 was conservatively used in the calculation. The maximum temperature difference is around 57°C which is one half of the spallation limit, 114°C .

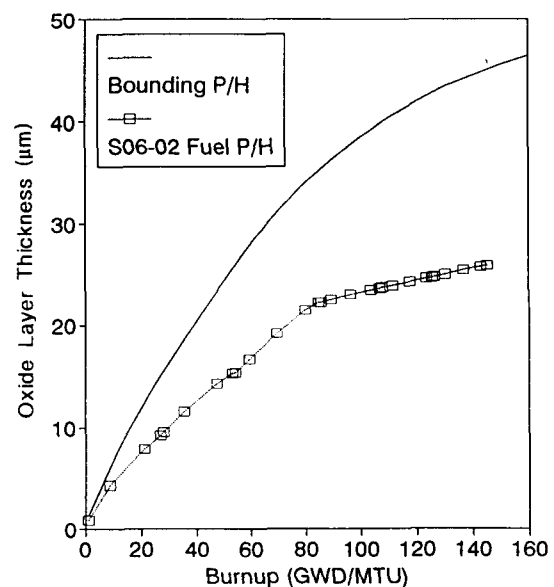


Fig. 4. Predicted Oxide Layer Thickness of the KMRR Fuel Cladding

Table 5. Thermal-hydraulic Condition of the KMRR Core

Variable	Value
Total Power (MW)	27.5
Coolant Pressure (MPa)	0.2-0.4
Coolant Mass Flow Rate (kg/s)	653.0
Average Coolant Velocity (m/s)	7.2
Core Inlet Temperature (°C)	35
Core Outlet Temperature (°C)	45
Coolant pH	5.5-6.5
Average Linear Heat Rate (kW/m)	42
Maximum Linear Heat Rate (kW/m)	104
Average Surface Heat Flux (MW/m ²)	1.27
Maximum Surface Heat Flux (MW/m ²)	3.15

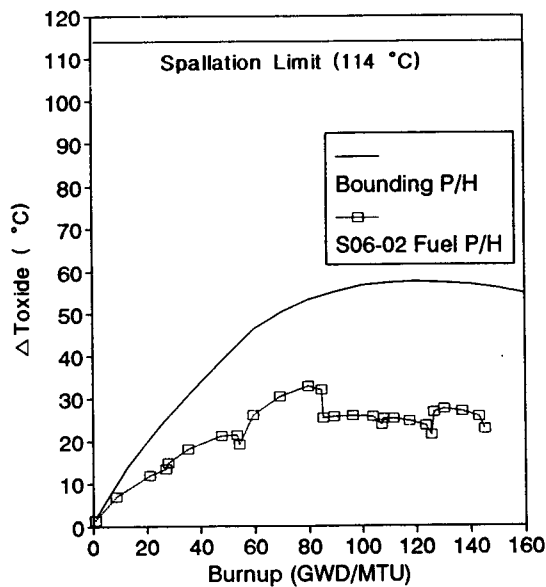


Fig. 5. Temperature Difference Through the Oxide Layer of the KMRR Fuel Cladding

even for the bounding power history. Therefore, it can be said that corrosion of the cladding will not result in the spallation of the oxide layer and subsequently will not impair the integrity of the KMRR fuel.

4. Conclusion

Through a literature survey and analysis of the corrosion data and corrosion correlations of the aluminum alloys, a modified Griess correlation for the aluminum alloy corrosion which can take into account the additional effect of the heat flux upon the aluminum alloy corrosion was derived and a limited validation of its application to the KMRR was shown. Then, the modified Griess correlation was applied to the evaluation of the corrosion of the KMRR fuel cladding. Evaluation was performed for the KMRR fuel with a bounding power history. Results showed that the cladding corrosion of the KMRR fuel even with a bounding power history satisfied the design criterion of the oxide spallation limit with a factor of 2 margin. Nevertheless, the applicability of the modified Griess correlation to the KMRR may need to be further verified through the KMRR fuel corrosion surveillance.

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