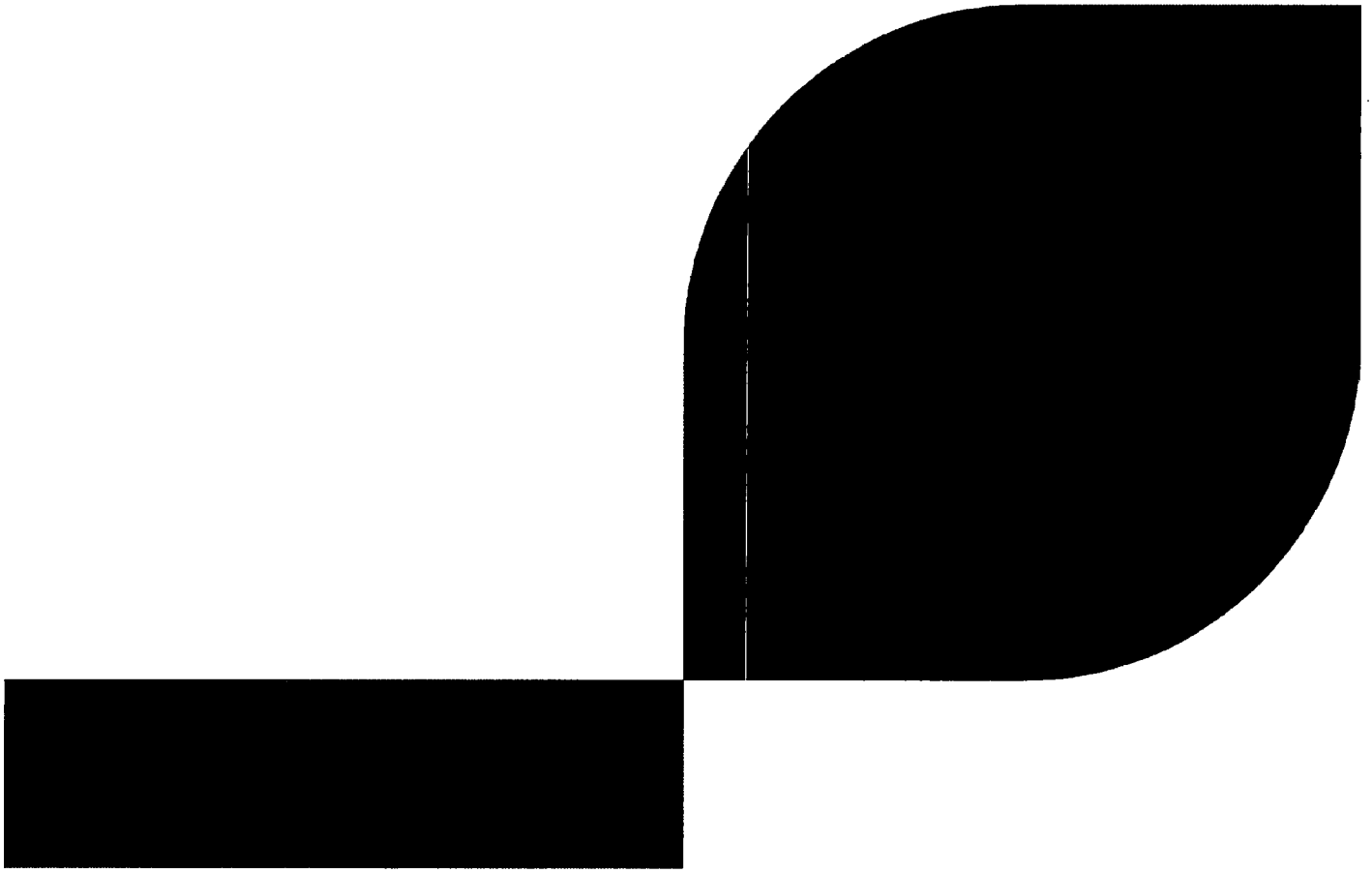

BAW-10247PA
Revision 0
Supplement 1Q2NP
Revision 0

# Realistic Thermal-Mechanical Fuel Rod Methodology for Boiling Water Reactors Supplement 1: Qualification of RODEX4 for Recrystallized Zircaloy-2 Cladding

## Responses to NRC Request for Additional Information

November 2013

**AREVA NP Inc.**

AREVA

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## Nature of Changes

| Item | Page | Description and Justification |
|---|---|---|
| 1. | | This is a new document. |

## Contents



## Tables



## Figures



*This document contains a total of 28 pages.*

# Response to NRC Request for Additional Information – BAW-10247PA Revision 0 Supplement 1P Revision 0

## Question 1

The following relates to cladding corrosion and hydriding.

## Question 1a

*Please provide the individual corrosion data along with cladding temperature (radial and axial location and fluence/time) and RXA Zr-2 model prediction and identify the fuel design the data was taken from. Also, include the maximum power level and burnup for each of the operating cycles for each rod. Do any of these data come from plants with power uprates? If so, please identify the data along with percentage uprate in power along with uprated core average power. This will help assess whether the data is applicable to today's fuel designs and operating envelopes.*

## Response 1a

Table 1 provides the requested information of the oxide database, with exposure and fast fluence being the local values at the location where the maximum oxide thickness was measured (of course, irradiation time is a global parameter). The data come from AREVA fuel irradiated in European reactors with annual cycle fuel management by selecting for each fuel rod examined pool-side by EC (Eddy Current) probe technique, the maximum value of the several EC probe linear scans usually performed on each rod. The irradiation lifetime covers the two-year cycle fuel management that is used in US, with up to eight years of operation and the exposures conservatively bound the [ ] limit approved for RODEX4.

Radial and axial cladding temperatures for each data point are not provided in Table 1. There is only minimal axial variation of coolant and cladding outer temperatures in a BWR coolant channel. This implies, as experimentally confirmed, that little axial variation exists with regard to oxide thickness, except the first 10% of the rod length at the bottom inlet region for which low coolant temperature and power levels exist through irradiation lifetime. [

]

Cladding outside temperature has little variation along the fuel rod length because the coolant temperature does not vary significantly axially and the film heat transfer coefficient increases with elevation as nucleate boiling regime is established by the increasing coolant temperature. Typically, coolant inlet temperature is approximately [

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[ ] Parametric studies for ATRIUM-10 and ATRIUM 10XM fuel were performed that resulted in a non-linear correlation of cladding outer temperature with linear heat generation rate. Figure 1 below illustrates the variation of cladding outer temperature with LHGR as described by the above relationship. It can be concluded that cladding outer temperature varies very little with axial location and linear heat generation rate. Therefore, the cladding outer temperature can be considered the same for any power and burnup values in the BWR normal operational domain. This includes power levels up to and including extended power uprates in U.S. plants.

**Table 1: RXA Zircaloy-2 Lift-off Oxidation Database Data**

[ ]

**Table 1: RXA Zircaloy-2 Lift-off Oxidation Database Data (continued)**

**Table 1: RXA Zircaloy-2 Lift-off Oxidation Database Data (continued)**

**Table 1: RXA Zircaloy-2 Lift-off Oxidation Database Data (continued)**

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**Table 1: RXA Zircaloy-2 Lift-off Oxidation Database Data (continued)**

**Table 1: RXA Zircaloy-2 Lift-off Oxidation Database Data (continued)**

[redacted]

**Figure 1: Outer Cladding Temperature Dependence on Rod Power**

**Question 1b**

*No model was provided for hydrogen pickup of RXA Zircaloy-2. NRC has developed cladding embrittlement criteria for loss-of coolant accident (LOCA) and reactivity initiated accident (RIA) based on hydrogen content. Does AREVA intend to include a hydrogen pickup model for accident analyses? If so, does AREVA intend to submit hydrogen pickup models for RXA Zircaloy-2 and CWSRA Zircaloy-2 as part of this review, also please provide supporting hydrogen concentration data for these models.*

**Response 1b**

**Background**

AREVA has submitted a hydrogen pick-up (HPU) model as part of the RODEX4 Thermal Mechanical Code Licensing Technical Report (LTR) (Reference 1). The U. S. Nuclear Regulatory Commission (NRC) judged that the database was insufficient to allow approval of this model, and it was therefore excluded from the Safety Evaluation Report (SER). Subsequently, AREVA has established an enlarged hydrogen pick-up database from new hot cell examinations to determine hydrogen pick-up levels of liner cladding consisting of RXA Zircaloy-2 material, from fuel rods irradiated in European power reactors. In US, AREVA uses Zircaloy-2 in the CWSR condition. The equivalence of the two separate lift-off based oxidation databases for the RXA and CWSR metallurgical conditions support the assertion that corrosion of Zircaloy-2 is the same regardless of the metallurgical state. Because hydrogen uptake is intimately linked to oxidation, hydrogen uptake is also considered equivalent for the two metallurgical states of Zircaloy-2.

The currently available industry data are typically presented as a function of burnup (BU), which is roughly proportional to the correct independent variable, namely irradiation time. The hydrogen is created from the oxidation reaction between the zircaloy cladding and the coolant, with a certain percentage, the hydrogen pick-up factor (HPUF), being absorbed by the clad and leading to the final hydrogen wppm content in the clad. The oxidation rate is also believed to be enhanced at high burnup. This HPU evolution is typically illustrated as a plot of hydrogen content vs. BU, which exhibits an accelerating level of hydrogen content with burnup. Typical hydrogen values at end-of-life BU (at ~62 GWd/MTU rod average BU) that were reported in the past ranged up to [ ] and exhibited a great deal of scatter [

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**Outline of H uptake mechanisms**

The oxidation of and H absorption by cladding is a very complex, thermo-mechanical, electro-chemical and diffusion-reaction process. The outcome depends on:

- The radio-chemistry of the coolant condition;
- The oxidative or reducing state of the coolant, which influences:
- The adsorption reactions at the oxide surface;
- The diffusion through the oxide of O ions and e-, as influenced by the O vacancies and the effect of second-phase precipitates (SPPs);
- The diffusion-reaction inside the metal which is influenced by temperature, concentration and stress gradients;
- The mechanical damage of the oxide, due to stresses created by the density mismatch between the oxide and the metal substrate.

It is extremely challenging to model this complex process in all its details, not only because of the physical and mathematical complications, but because of incomplete knowledge of the mechanisms involved. Therefore, a simplified approach is proposed, which is also what has been adopted in the past on the basis of the experimental data and research into this matter.

The whole body of data accumulated in more than 50 years of investigations on zircaloy alloy corrosion and H pick-up, led to the general conclusion that there is a strong link between the oxide layer thickness and the H uptake, hence the use of HPUF was and still is widespread. However, the exact value of HPUF is dependent on the environmental conditions, such as water chemistry and also on material properties and composition.

In spite of extensive research the exact mechanisms involved in H ingress into the cladding are yet to be identified. A research program is currently underway in the NFIR program [

]

**Role of [ ] and liner as H sink**

Due to hydrogen's negative migration energy in zircaloy, H migrates down the temperature gradient (from highest to lowest temperature regions). In operation at typical high power levels, a radial temperature gradient of 20 to 35 K exists within the cladding with the cool end at the cladding outer surface. Therefore, in those conditions, the temperature gradient opposes but

doesn't prevent H ingress into the cladding and a dynamic equilibrium is established between the concentration gradient (higher concentration at the cladding outer surface) and the temperature gradient. In addition, after the solubility limit is attained at the cladding outer surface, a hydride layer can develop there.

During periods of time with low Linear Heat Generation Rate (LHGR) [ ] the temperature gradient is very small and therefore the inward H diffusion is greatly enhanced. This effect of the heat flux on H pick-up [

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This specific feature of liner being a sink for the H taken up by the cladding is the result of the lower H solubility limit of the more pure zirconium liner, which creates a driving force for H migration to the zirconium liner at the cladding inner surface. Opposing this liner sink effect, there are two other forces, namely the concentration gradient and the temperature gradient.

[

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This liner sink effect is well documented for both BWRs and pressurized water reactors (PWRs). The PWR example is documented in Reference 3. For BWRs, recent studies in the frame [ ] clearly showed that a significant [ ] portion of the H intake is located in the liner (Reference 4).

Based on the theoretical arguments and experimental evidence for the liner sink effect presented above it is clear that the liner enhances hydrogen uptake and therefore, hydrogen uptake of liner cladding bounds the hydrogen uptake of non-liner cladding.

[

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Support for this correlation to the power history type can be found in the data published by another fuel supplier [

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(Reference 5). Also, data reported by the Japanese Atomic Energy Research Institute (JAERI) in the context of Nuclear Safety Research Reactor (NSRR) Reactivity Initiated Accident (RIA)-type studies showed the impact of low power at the end of the irradiation lifetime (Reference 6).

A number of papers have proposed that hydrogen pickup acceleration at high burnup can be explained by SPP's dissolution. In Reference 7, complete SPP dissolution is assumed to be the threshold for an exponential acceleration of hydrogen uptake. Before that, hydrogen uptake is postulated to occur by H diffusion through SPP's by short-circuiting the barrier layer at the base of the oxide film, in which H diffuses very slowly. On the contrary, at high fluences, it is postulated that complete dissolution of SPP's causes deterioration of the barrier layer which leads to both accelerated hydrogen uptake and oxidation. However, no mechanism is proposed for this deterioration of the barrier layer by complete SPP dissolution, which somehow becomes permeable to H.

The kinetics of SPP dissolution is still under investigation, but general trends have been identified, albeit with some differences between material variants and studies. In all studies reported in the literature for Zircaloy-2, Zintl (Fe-Ni precipitates) SPP's remain crystalline and dissolve more slowly than the Laves (Fe-Cr precipitates) SPP's. A rapid decrease of the small Laves SPP's during the first cycle (corresponding to ~ 1-2 E+21 n/cm2) is followed by a very slow decrease up to terminal fast fluences of the operational range (Reference 8).

Reference 9 also confirms the rapid dissolution of small Laves SPP's (less than 30 nm and less than 70 nm at very high fast fluence), nevertheless an important fraction of both large Laves and Zintl SPP's remain even at very high fast fluence. Alternatively this reference suggested another apparent correlation, namely of accelerated hydrogen uptake with Fe depletion from the SPP's by their dissolution and decrease in number, but again it is stated that "the exact physical mechanism is not yet understood".

The apparent correlation between hydrogen uptake and metal microstructure evolution with exposure (fast fluence), in particular the dissolution of SPP's does not appear to be a causal relationship and no mechanism was proposed, let alone proven to substantiate such a causal correlation. The absence of a direct causal correlation between SPP dissolution and increased hydrogen uptake is supported by counter examples provided by the AREVA HPU database presented herein. [

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This is further supported by the corrosion behavior of structural materials, which operate without any temperature gradient. As reported in Reference 10, samples of Zry-2 and a developmental high Fe Zry-2 material were irradiated in the coolant of a commercial BWR and afterwards examined in the hot cell. It was found that SPP's did not disappear after 6 full cycles. However, the hydrogen uptake pickup fraction increased after 4 cycles until the final sixth cycle of irradiation; although the paper does not credit an acceleration of oxidation after 4 cycles, the data presented in Figure 4 of Reference 10 shows accelerated oxidation during cycles 4-6.

Support for the link between enhanced hydrogen uptake for thick oxide films (thick film hypothesis mentioned below) in conjunction with zero thermal gradient (low LHGR equivalent) conditions is furnished by Reference 11: Zry-2 coupons with low oxide thickness displayed low HPU, while large HPU was associated with thick oxide coupons. Although a speculative correlation to fast fluence is mentioned in Reference 11, it is clear that the data support the thick film threshold as a condition for enhanced HPU, concomitant with low heat flux.

**H uptake model**

Therefore, the model described herein relies on the following three main features, which act synergistically at long irradiation times and could result in enhanced HPU in those conditions:

[ ]

The first and third items have been described in the previous section, while the second one is presented below.

Although no generally accepted model exists that could explain all aspects of Zry-2 corrosion and H pick-up, a widely accepted phenomenological feature of Zircaloy-2 corrosion is the so-called "thick film" hypothesis (Reference 12), which asserts that enhanced corrosion and hydrogen uptake occur in thicker oxide layers. It is remarkable that both oxidation and HPU are increased after the oxide thickness exceeds the thick film threshold. The explanation proposed for this seeming paradox is the mechanical damage and crack tunnel formation in the outer thicker oxide layer. In addition, the thick film hypothesis can be correlated with the Ni segregation on the grain boundaries as a result of SPP dissolution, which occurs after a certain irradiation time.

The thick film hypothesis was confirmed in a number of studies, such as References 10-12, where the accelerated oxidation and/or hydrogen uptake was noticed at about the same oxide thickness, namely 11-13 microns.

The inner 2-3 micron-thick layer which is tetragonal zirconia was found to be very opaque to H, i.e., H diffusion through it is extremely slow if anything (Reference 12). Temporary easier access routes though oxide pores and/or SPP crossing the oxide-metal boundary are the most likely explanations for the thick film behavior. In addition, at long irradiation time, the SPPs are dissolved and contribute to the decoration of oxide grain boundaries by alloying elements, which become shortcut pathways for H ingress.

[

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When LHGR decreases during the second part of the irradiation lifetime, the temperature gradient practically vanishes and the surface hydride layer dissolves leading to H transport into the metal. Further H ingress is then possible (assisted by the liner sink effect of the liner material). This leads to increased H uptake.

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[ ]

In addition, the change to the higher HPUF values of F2 is conditioned by exceeding an incubation time threshold for low power operation that was set to:

[ ]

The power thresholds defined above are applicable to other fuel designs because the temperature drop across the cladding is similar for all fuel designs as the ratio of thickness to mean radius has been kept approximately the same for all fuel designs. Indeed, the temperature drop across the cladding thickness, t, can easily be calculated, using a constant zircaloy conductivity, k, and equating the heat flux at the cladding outer surface, R from Fourier's law with the expression derived from the definition of the linear power, P, to obtain:

$$\Delta T = \frac{P}{2\pi k}\frac{t}{R}$$

The uniform oxidation model is a pre-requisite for calculating the hydrogen uptake by the above formulas. The hot-cell examinations that produced the HPU database also include metallographic determinations of the average uniform oxide at the neighboring location to the cut used to determine hydrogen uptake by a high-vacuum fusion and chromatographic measurement of hydrogen.

The model is based on the MATPRO (Reference 13) model with a variable adjusting factor that was used for calibration on the uniform oxide database. The oxide thickness is calculated with this variant of the MATPRO model, so that best-estimate agreement is obtained in the high

oxide end of the database, [ ] To that end a variable adjusting factor was implemented. The increase of oxidation rate above a certain oxide thickness is in accord with the thick oxide hypothesis adopted as a model assumption. The following values have been determined by calibration:

[

]

**H model calibration**

The results of benchmarking on the AREVA HPU for Zry-2 are presented below.

The model was calibrated based on the current enlarged dataset, consisting of [ ] by using the re-calibrated uniform corrosion model for Zry-2 as defined above. The results obtained are displayed in Figure 1. The calibration goal was to achieve a best-estimate model, especially in the high-end range; if necessary a moderately conservative, over predictive model is acceptable in the low-end domain so that over [ ] H pick-up range a best-estimate model is obtained.

The results of the re-calibrated uniform oxidation model are presented in Figure 2 and show good centered prediction for the whole database domain. Figure 3 overlays uniform oxide calculations and measurements against irradiation time and shows the slight over prediction of the low end of the measured oxide range.

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**Figure 2: Calculated and Measured Hydrogen Uptake**

**Figure 3: Calculated and Measured Uniform Oxide Thickness**

**Figure 4: Calculated and Measured Uniform Oxide Thickness vs. Time**

**Figure 5: Calculated and Measured HPU vs. Time**

**Figure 6: Two Typical High Power Histories and Associated Low Measured HPU**

**Figure 7: Two Power Histories with Extended Final Low Power Period and Associated High Measured HPU**

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