A NUMERICAL COMPONENT TO EVALUATE THE IODINE CORROSION IN A NUCLEAR FUEL ACCOUNTING FOR CHEMISORPTION MECHANISMS ZIRCONIA LAYER IMPACT ON SCC.

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Abstract. In PWR plants, nuclear fuel pellets (cylinders 1.2 cm length, 0.84 cm diameter) are piled in nuclear fuel rods placed in assemblies, each containing 265 fuel rods. the fuel rod is a tube manufactured in a zirconium alloyed material. Fuel cylinders are piled to form a fuel stack shorter than the overall tube length to maintain a remaining upper volume called plenum, able to allow fuel stack elongation and accommodation of the gaseous and volatile fission products released during operation. The fuel stack (3.6-4 m depending on the design) is typically maintained during handling operation by a spring placed in the plenum. The fuel rod is closed at both ends by sealed end plugs after filling dead volumes with a neutral gas (helium) with a pressure ranging from 1 to 34.5 bars. The cladding insures the first confinement barrier against radionuclides dispersion. During severe power transients some fission products, like iodine, are expected to assist cracks initiation and propagation at the inner surface of the cladding. Our major concern here aims improving understanding the involved mechanisms, accounting for the zirconia layer able to develop as soon as a closed contact is locally established between the pellet fragments and the cladding inner surface.

In this work, we extend a specific numerical component, developed by EDF, coupling two numerical software codes [1]. The first one aims simulating the evolution of the fuel isotopic composition and the second one aims evaluating the fuel dioxide chemistry. The growth of the zirconia layer has been modelled and integrated in the numerical tool [2]. The scenario of Stress Corrosion Cracking (SCC) previously defined has been improved to consider the full impact of the zirconia layer on SCC. The role of the zirconia layer can be mechanical, thermal or even chemical. Assumptions and coupling of the phenomenon are discussed.

To make demonstration, a sensitivity analysis is performed. This study considers several

zirconia layer thickness for two cases: a high burn-up fuel rod operated in normal PWR conditions, and a fuel rod segment pre-irradiated in a power plant and then submitted to an incidental power transient simulation in a MTR. The results demonstrated a protective effect of the zirconia layer. As a consequence, the corrosive attack of the clad is delayed. The logarithm function of the iodine chemisorption kinetic appears to be proportional to the zirconia thickness. The simulation analysis validates the extension of the SCC scenario. The results are in good agreement with experimental observations.

1   INTRODUCTION

The numerical component NUAGES – Numerical Unit ensuring ANGE coupling with EDMOND and STRAP – is an embedded software structure "like russian dolls ", in which the "father" component call and plug three "son" components [Fig 1]-[1,2].
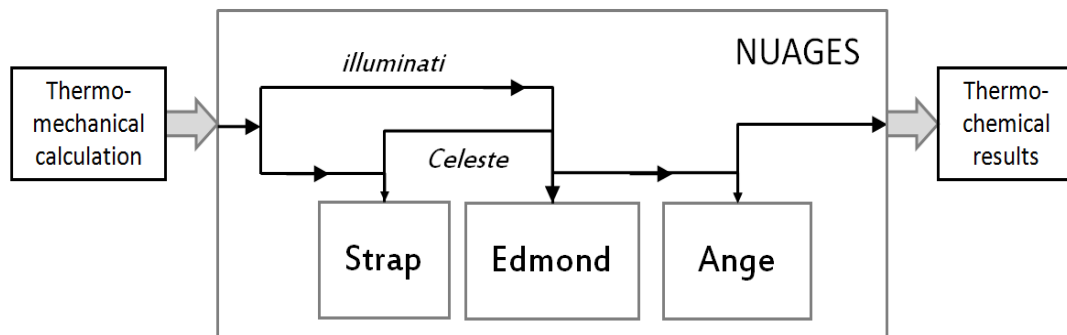

Figure 1 - Scheme of a calculation performed with NUAGES [3,4]

Version 1.1 of the component includes a scenario for modelling thermochemistry processes of the cladding Stress Constraint Corrosion (SCC). In parallel, it was developed to enable an estimation of the local zirconia layer growth at pellet cladding interface, and of the ballistic drop of fission products ejected from the pellet toward the cladding as well [5,6].

Bibliographical studies based on the analysis of a large number of destructive post ramp examinations, enlighten that the zirconia could have a protective effect against the SCC cracks propagations. The numerical component NUAGES, allows in a relatively simplified way modeling both phenomena and the way they could be coupled. We are going to present more in detailed the numerical tool developed and then show how results are quite promising regarding the prediction of the observations reported in literature [7,8].

2   PROTECTIVE EFFECT OF ZIRCONIA LAYER ON SCC

A very large number of post ramp examinations were conducted in the past and are still pursued to better understand and simulate the PCMI risk of failure during severe power transients. At this stage, most of observations lead to some main conclusions on which engineering studies agree:

1. The presence of oxygen in small quantity in the neighborhood accelerates SCC processes. It was evidenced by three tests: under air atmosphere, under inert gas atmosphere to lower the Oxygen potential and in presence of warmed shavings of zirconium (the oxidation of which lowers the partial pressure of oxygen with values close to those evaluated in the fuel rod) [9].

2. Presence of oxygen in high quantity in the test environment inhibits the reaction with the iodine. This was shown from local analyses realized on a surface of zirconium free of constraints, submitted to a molecular flow of iodine. The study concerns the impact of a flow of oxygen managed in the same place as the flow of iodine. It shows that there is a competition between the iodine and the zirconium chemical reactions on one hand, and the iodine and the oxygen chemical reactions on the other hand [10, 11].

3. Presence of a pre-oxidation layer important (upper to 0.2μm thickness) on the inner surface of the metal is beneficial; nevertheless this layer can break easily with the differential strain, letting the iodine reach locally the surface. Tests on pre-oxidized claddings either in an oven or before fabrication, show that a too thin layer, like the one naturally formed in ambient atmosphere (10-15 nm), has no protective effect on the metal [9].

Eric Walle proposed some additional experiments to specify the role of the oxygen during SCC [12]. Samples used are leaky rings (10 mm in diameter) of recrystallized Zircaloy-4. A strain control loading path is applied in non–irradiated conditions. As there is no additional mechanical stress on the sample, the loading is only representative of the crack initiation mechanism (and not for propagation). The tests differ by the oxygen pressure, which is controlled just before the test, when the sample is sealed in a glass tube. They also differ either from the presence (or not) of a pre-oxidized layer on the cladding inner surface, either from the test duration. The results seem to indicate:

4. That crack initiation is due to mechanics. On pre-oxidized samples, perpendicular cracks in the interface are developing in front of the oxidation forehead. The authors consider thus reasonable to think that the combined effect of the formation of oxide and the mechanical loading are at the origin of a purely mechanical initiation of cracks in the metal.

5. That the presence of oxygen localizes the phenomenon of SCC. Without oxygen we observe a generalized corrosion of the samples, while the presence of oxygen allows the localization of the SCC [Fig. 2]. The protection of the surface by a pre-oxidized layer of 1 or 10μm turns out to be a sufficient (but not necessary) condition for a localization of the chemical attack relative to SCC.

6. That the oxygen can lead to a passivation of the metal if the quantity is important enough so that the formation of an oxide layer on the metal surface is faster than the attack by the iodine [Fig 2].
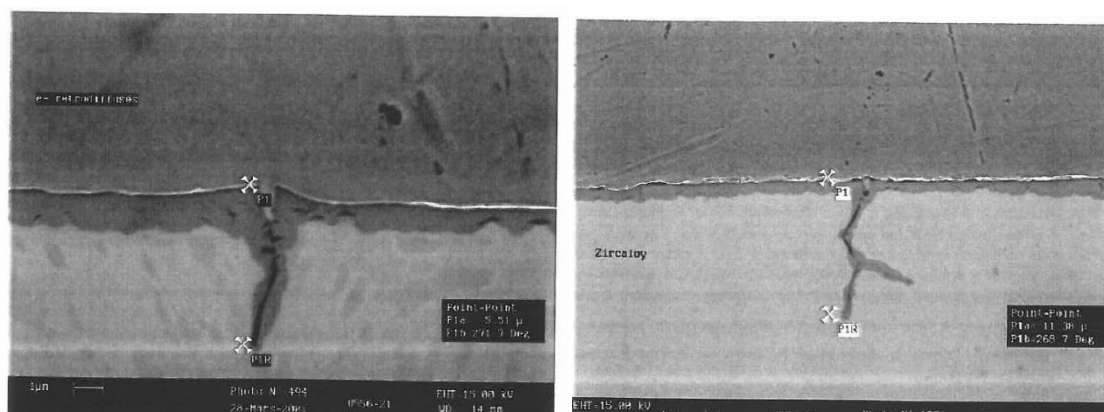
Figure 2 : Cross section of two samples, *in* [12]

The sample on the left is a reference that has been preoxydized at 1 μm, without iodine, and submitted to a 500 hour testing. The sample on the right has been preoxydized at 1 μm, with a partial pressure of oxygen of 0.01 bars at ambient temperature, the a diiodine partial pressure of 0.01 bars and finally submitted to a 150 hour testing.

3 EXTENDED MODELING SCENARIO OF SCC

Here are the main steps and assumptions for the modeling scenario retained for SCC without considering the impact of the zirconia layer [7,8]:

Step 1. Inventory of the neutronic balance sheet

Step 2. Ballistic migration and drop of fissions products through the pellet-cladding interface

Step 3. Thermochemistry: evaluation of condensed compounds and gaseous species (like I, I_2, CsI O_2) and also their chemical potential.

Step 4. Evaluation of corrosive source term

First assumptions. Fission gas released will be consider either from a conservative point of view (100% of fission gas is instantaneously released), either by means of a best estimate evaluation (the fission gas released rate is predicted by a thermo-mechanical code).

Step 5. Corrosive attack of the cladding

Second assumption: Cracking initiation is due to the mechanical loading (the corrosion only assists the failure mechanism)

Third assumptions: The corrosive attack is due to gaseous iodine. The monoatomic specie has been preferred to the diatomic one.

Fourth assumption: Because of lack of evidences, an hypothetical radiolysis of gaseous species is not considered. As a consequence gaseous CsI does not dissociate.

The main result of this scenario consists in an evaluation of the iodine and oxygen deposit rate on a zirconium crystal due to chemisorption. Indeed chemisorption is the first step for metallic corrosion.
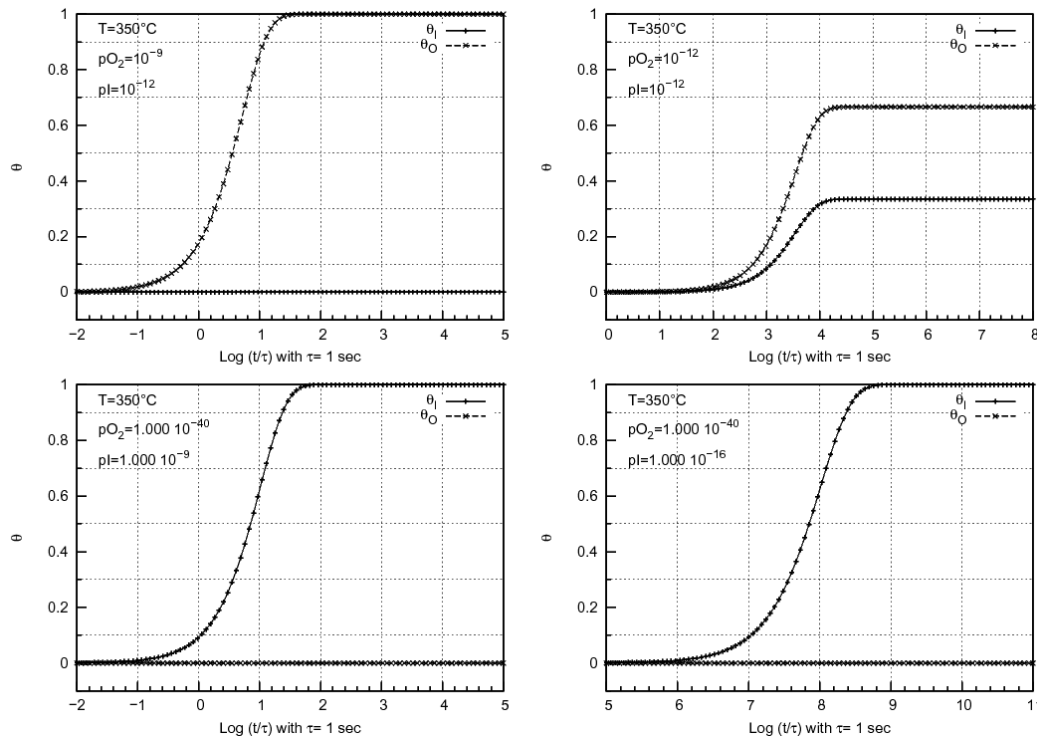

Figure 3. Chemisorption examples curves [13]

Figure 3 shows examples of chemisorption curves which can be obtained at the end of SCC modeling. Those of the top correspond to two virtual cases where the oxygen is either in excess compared with iodine, or in equivalent quantity. Both bottom graphs correspond to two cases presented in Eric Walle's pHD thesis [12], the one on the left would be representative of a rod at the end of its life and the one on the right of a rod submitted to a power transient.

Let us now consider the role of the zirconia layer:

From a mechanical point of view, the zirconia is known for its very low mechanical resistance, even if the literature does not mention very homogeneous quantitative data on the subject [14,15]. As far as the previous study led on the SCC assume mechanical cracking initiation process, the wisest choice is to adopt a conservative approach:

Fifth assumption: No mechanical resistance of the zirconia layer.
In case of a power transient, we consider that the zirconia layer is systematically broken.

So, according to this assumption, zirconia will have no mechanical protective role towards SCC. The latter could be only thermal or chemical.

From a thermal point of view, zirconia has a lower thermal conductivity than zicaloy. As a result, the thermal gradient in the cladding is modified. Considering that the thickness of the zirconia layer is low compared to the cladding thickness (ratio of the order of 1,5 %), it is likely that the modification of the thermal gradient only affects the vicinity of the pellet-cladding interface. Nevertheless, it cannot be neglect because it is exactly at this location that the SCC (known as a localized phenomenon), happens.

This impact was taken into account in a new version of NUAGES [1.2]. Modelling considers a thermal conductivity of 15 W/m/K in the cladding, and 1.5 W/m/K in the zirconia layer (so a ratio of 10 between the two materials).
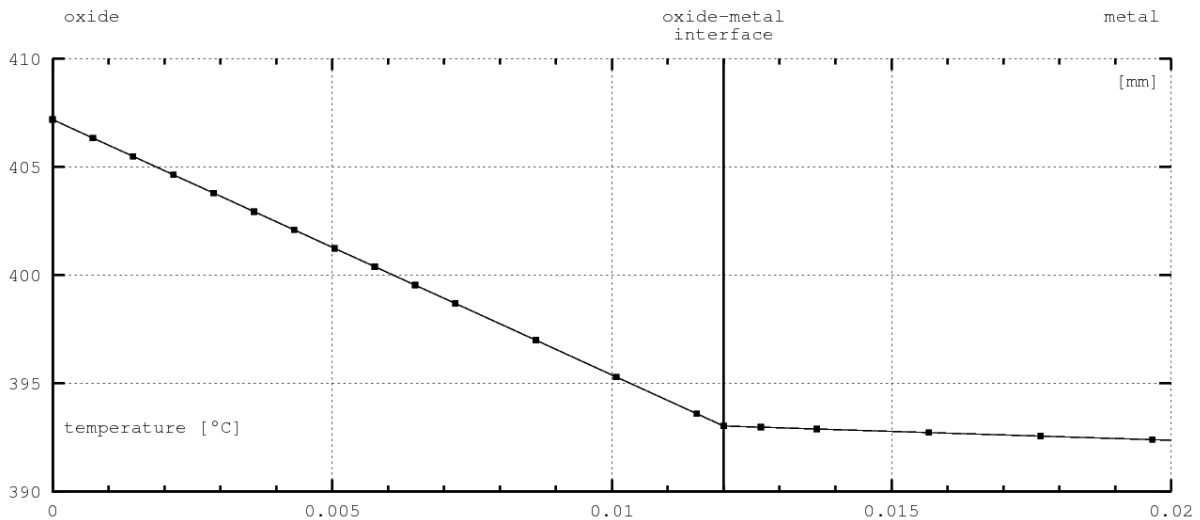

Figure 4 – Example of a thermal gradient, in a rod submitted to a power transient, considering a 12 μm thickness of zirconia at the inner surface of the cladding [16].

Figure 4 shows an example of thermal gradient considered in NUAGES v1.2, for a rod and a zirconia layer thickness that favor a huge thermal impact. The temperature drop through the zirconia layer is in this case about 14°C. We assume in the same time a systematic presence of crack in the zirconia layer. The temperature to be considered for chemisorption is then the temperature at the oxide-metal interface. This temperature is slightly lower than the temperature at the zirconia layer surface. The temperature drop in the zirconia layer is nevertheless low to explain a protective effect in absence of crack initiation.

From a chemical point of view, the presence of zirconia results from an oxygen flow from the fuel oxide toward the cladding. The oxygen is taken in the periphery of the pellet. Logically, the oxygen potential decreases. As a consequence, the chemical balances are modified. In other words, for a given element (iodine for example) the molar quantity is not modified, but chemical forms proportions between I(g) I_2 (g) or CsI (g) are modified.

Such effect is directly taken into account in NUAGES, in the sequence between EDMOND and ANGE [Fig 1]. The only constrain is to start calculations with a positive zirconia thickness and to provide an adequate time increment in the dataset file for the zirconia growth modeling [5].

4 CALCULATIONS DEALING WITH THE PROTECTIVE ROLE OF ZIRCONIA ON SCC

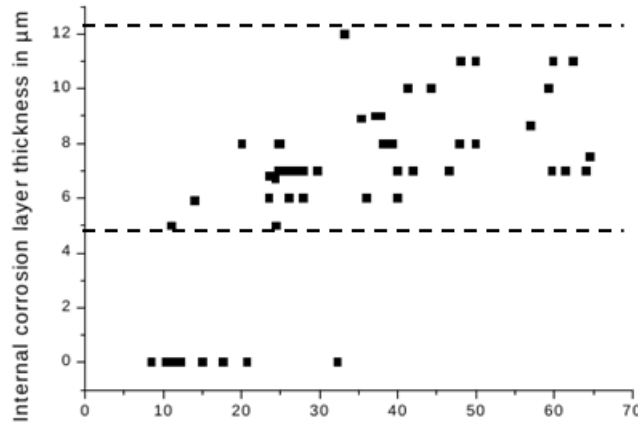Figure 5 presents the zirconia layer thickness that has been measured in fuel rods:

Figure 5 – Zirconia layer thickness, in [17,18]

As far as we know, there are no observations below 5 μm. The growth starts with localized zirconia islands, between which a continuous layer progressively develops. Studies let expect that this initiation phase is very quick, as soon as very punctual hard contacts happens between the pellet and the cladding. It is also observed that whatever the burn-up is, the zirconia layer growth is limited to about twelve micrometers. If we analyze more in details, for high burn-up the inner surface of the cladding penetrates HBS specific structure in the pellet rim, showing a wrinkled aspect. This phenomena strongly increases the specific surface between the cladding and the ceramic, making it difficult to measure the zirconia layer thickness [5].

For calculations, we choose to model the evolution of the observed thicknesses from 5 μm to 12 μm, with a 1 μm step. For demonstration, we used again the two reference calculations R_1 and R_2 rod, previously studied when validating SCC modeling scenario without taking into account any zirconia layer [8].

R_1 is a UO_2-Zy4 fuel rod irradiated during 5 cycles, to an average burn-up of 60 GWj/t (base irradiation)

R_2 is a power ramped segment taken from a fuel rod pre-irradiated during 3 cycles to an average burn-up of 45.3 GWd/t. This R_2 segment was conditioned at 195W/cm during 16h40. The conditioning rise was of 10 W/cm/mn. The power of the high level was of 466 W/cm. The average power was 470 W/cm. The power increase rate was of 100W/cm/mn. These conditions correspond to a severe power transient. Thus, this rod failed after 4mn04s. The holding time was of 1h34mn.

Each simulation was performed twice:

- Assuming 100 % fission gas release (conservative approach of SCC scenario),

- Providing a best-estimate evaluation of the fission gas release history (CYRANO3 calculations).

Finally, considering different zirconia layer thicknesses, we performed 36 simulations for
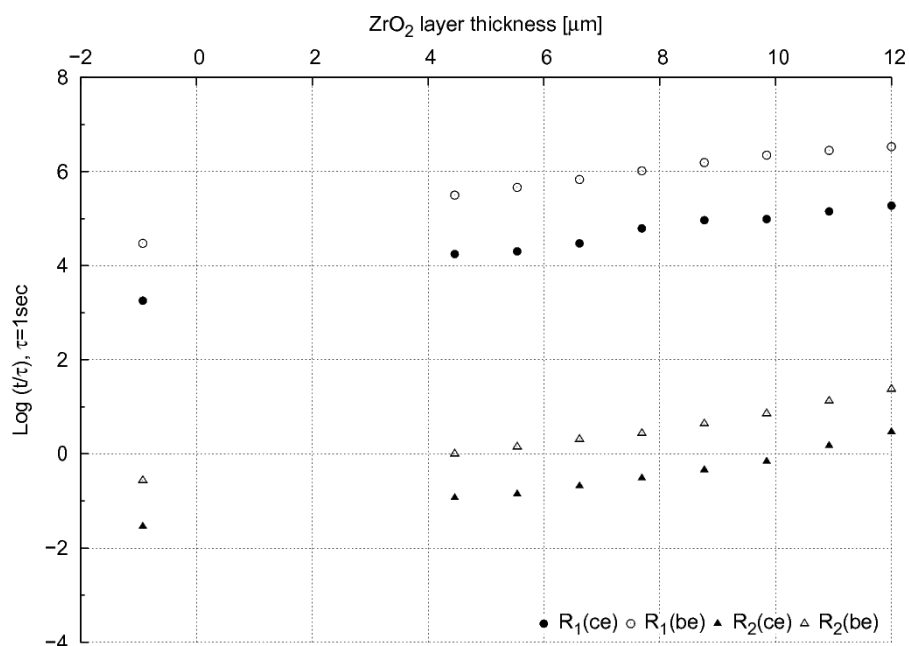
this sensitivity analyses.

Figure 6 – Zirconia impact on chemisorption kinetics, NUAGES v1.2

The covering rate of a zirconium monocrystal by iodine (to 1 more or less 10^{-6}) is delayed in presence of a zirconia layer. The delay is proportional to the layer thickness [Fig 6].

There is a linear relationship between zirconia layer thickness and the logarithm of delayed impact. Comparing the four cases here above, the slope of the linear relationship seems to be the same. Then, we manage to draw the results considering: $Log(t/\tau) - Log(t/\tau)_0$, $Log(t/\tau)_0$ standing for $Log(t/\tau)$ in case of a zero layer thickness [Fig. 7].

The linear relationship that quantifies the delayed impact of the zirconia layer on chemisorption can be then written:

$$Log(t/\tau) = Log(t/\tau)_0 + 0.167 e_{ZrO_2} - 0.044$$

with e_{ZrO_2} standing for zirconia thickness [μm] and τ=1sec.

The regression coefficient gives 0.897. It is quite satisfying. All the simulations points are quite uniformly distributed on each side of the regression line, at a maximum distance of twice the standard deviation. From a quantitative point of view, for a zirconia layer of 6 μm, the chemical reaction step is multiplied by ten. So far, this modelling confirmed the protective role of the zircon layer against SCC [Fig. 7].

We showed previously that the temperature drop in the zirconia layer was not enough to explain its protective effect. The modelling presented here suggests that the protective effect has a chemical explanation. Indeed, the decrease of the surrounding oxygen potential induces a decrease of the Iodine monoatomic gaseous potential, which is the corrosive specie retained

in SCC scenarios. We have now to check that the SCC modelling scenario remains valid.
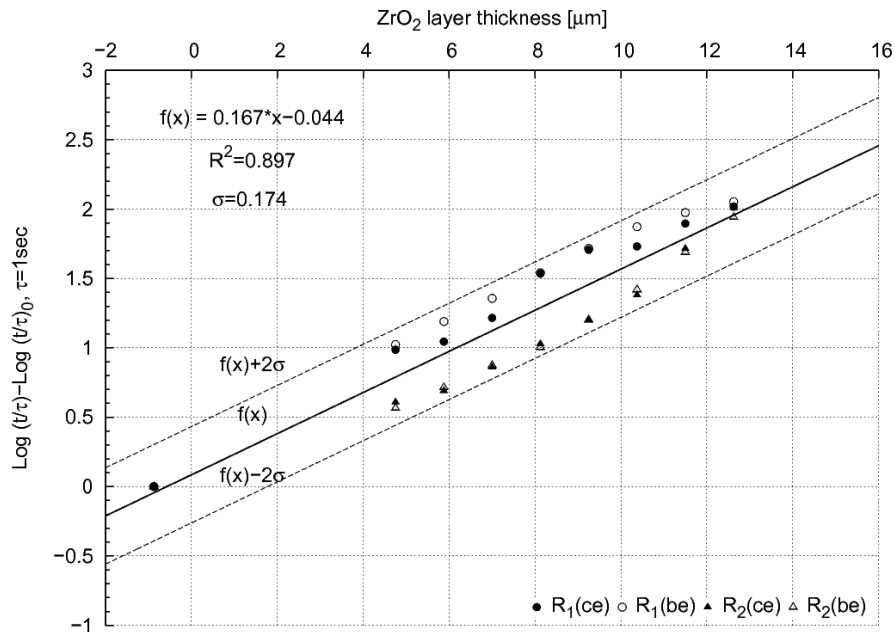

Figure 7 – Quantification of zirconia impact on chemisorption kinetics, NUAGES v1.2

5 VALIDATION OF SCC SCENARIO ASSUMPTIONS

The first assumption considers an upper bound and a lower bound to take into account, with very fast computing times, the fission gas release. We can observe on figure 6 that this two bounds do not prevent the differentiation of the high burn-up rod (R_1) and the rod submitted to a power transient (R_2). Whatever the assumption on fission gas released is, the chemisorption is always faster in the case of the power transient, with three orders of magnitude. As a consequence, the extension of SCC scenario accounting for the the zirconia layer does not modify assumptions relative to fission gas release. They are adequate to answer the industrial specification in term of simulation.
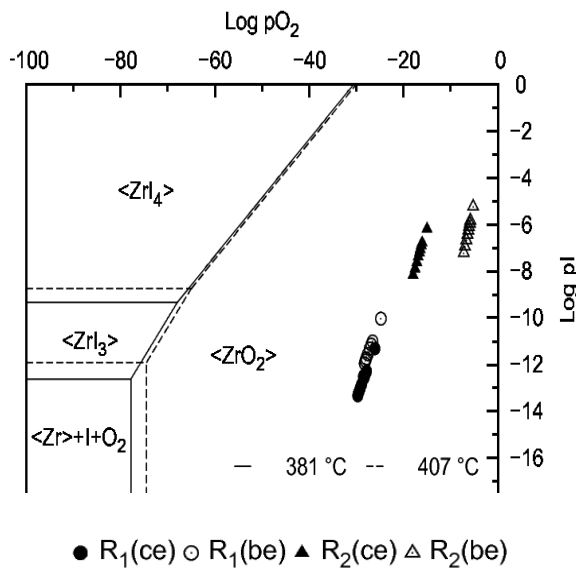
Figure 8 Zr-I-O binary diagram, NUAGES v1.2 built with equations available in [12]
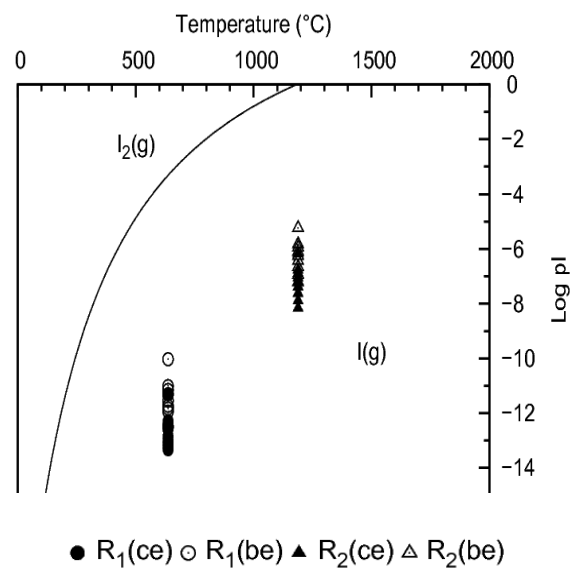
Figure 9 Vapor pressure of gaseous iodine, NUAGES v1.2, built with equations available in [19]

When equilibrium is established, the thermodynamic diagram predicts the development of zirconia at the inner surface of the cladding [Fig. 8]. This is in accordance with the in pile observations. On another hand the presence of a crack crossing the zirconia layer and reaching the zirconium interface, can lead to a steady-state thermodynamically compatible to initiate Iodine corrosion. In this case, even if the presence of the zirconia layer lowers the oxygen potential in the gap, it has no effect at the zirconia-zircaloy interface where ZrI_x species are formed. Therefore, the hypothesis of a crack mechanical initiation stays a necessary condition for the SCC modeling scenario.

The third assumption considers that diatomic gaseous iodine can be neglected compared with the monoatomic form. Figure 9 shows the proportions of monoatomic iodine vs diatomic iodine. Nevertheless gases are produced inside the pellet. Thus, we had to postpone on the diagram the partial pressures of iodine estimated by calculations, according to the average temperature of the pellet. Because it lowers the potential of gaseous monoatomic iodine for an equivalent temperature, the presence of zirconia does not call into question the predominance of this form against the gaseous diatomic iodine. On the contrary, it tends to strengthen it. The assumption considering the monoatomic Iodine form mainly responsible of the corrosive attack remains valid [Fig. 9].
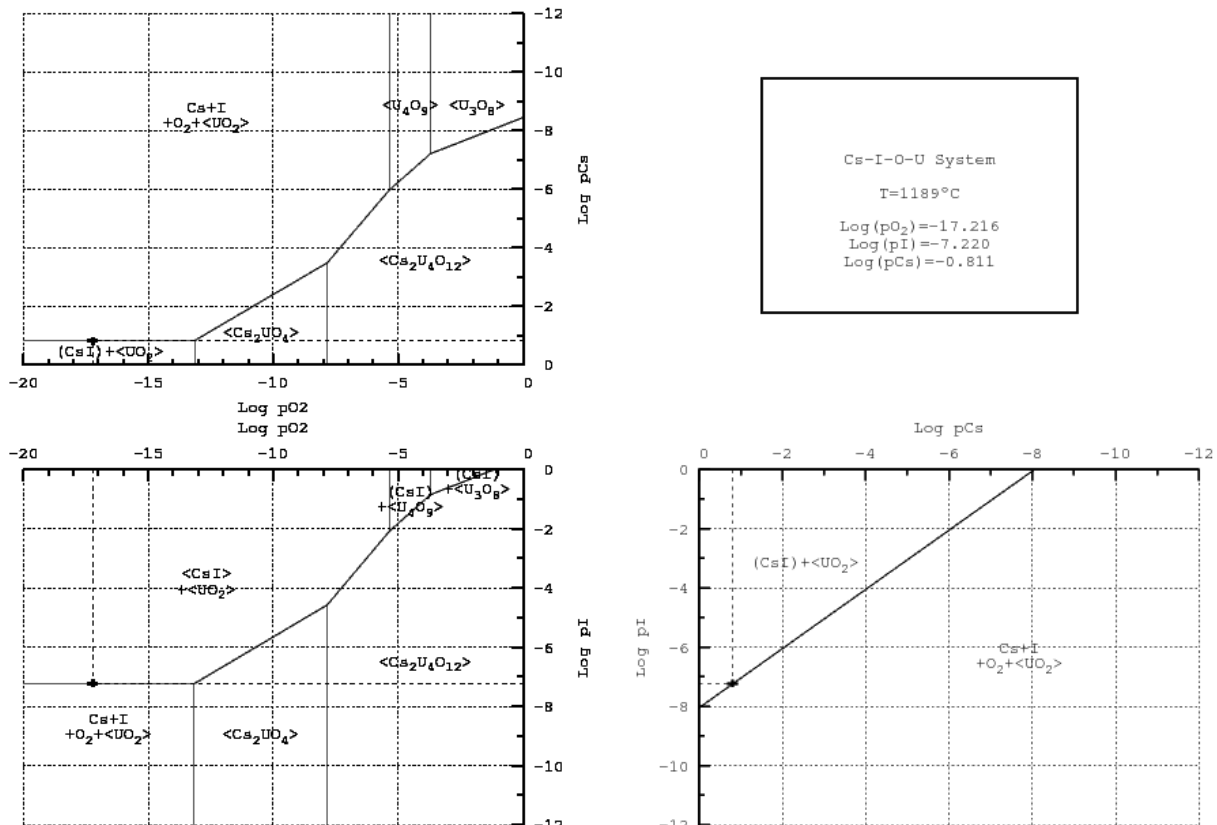
Figure 10 : Ternary Cs-I-O-U diagram, R2 calculation with a 12 μm zirconia layer thickness,

NUAGES v1.2

Finally, the fourth assumption considers the radiolysis phenomenon in pile as marginal. CsI(g) is not a corrosive gas for the cladding. However in case of dissociation, it would provide monatomic or diatomic iodine. CsI(g) is produced in large quantities within the pellet. Moreover, according to the thermodynamics diagram balance, the potential of cesium is widely higher than that those of iodine and oxygen. We also notice that the calculations are always situated in the balance between I (g) and CsI (g) [Fig. 10].

At first glance, the zirconia layer does not carry new information about the hypothetical role of radiolysis. But, we also can consider another point of view. Let's consider that radiolysis plays a significant role in pile. It would mean a large scale dissociation of CsI (g). As a consequence the potential of gaseous iodine species would be the same whatever is the zirconia layer thickness for a given burn-up. Calculations would not be able anymore to predict the protective role of the zirconia on SCC observed experimentally.

In conclusions coupling a protective effect of the zirconia layer to the CsC model, remains consistent with all assumptions underlying the industrial CSC model. It even strengthened the original model.

6   ADEQUACY BETWEEN SIMULATIONS AND EXPERIMENTAL OBSERVATIONS

We suggest here to confront experimental observations which describe the protective role of the zirconia towards SCC with our scenario and the results of simulations.

1.   Presence of oxygen in small quantity in the middle accelerates SCC The calculation consolidates this observation, the more the quantity of oxygen decreases the more the reaction mechanism of chemisorption of iodine is delayed [Fig. 6].

2.   Presence of oxygen in high quantity in the test environment inhibits the reaction with iodine. In our calculations, the presence of oxygen is always unimportant compared with the quantity of iodine. However, considering a hypothetical case in which the quantity of oxygen would be equivalent to the initial quantity of iodine [Fig. 3], the oxygen would coat zirconium. inhibiting reaction with iodine. The chemisorption model thus accredits the experimental observations.

3.   The presence of a pre-oxidation layer protects the metal if the thickness is self-important (upper than 200 nm). The one naturally formed in ambient atmosphere (10-15 nm), has no protective effect on the metal. Our simulations allowed to quantify the impact of zirconia towards SCC :

$$\text{if } e_{ZrO_2} = 0.2 \text{ µm, then } Log\left(t/\tau\right) - Log\left(t/\tau\right)_0 = -0.0116 \text{ and } Log\left(t/\tau\right) = 0.974 \, Log\left(t/\tau\right)_0$$

So, a 0.2 µm zirconia layer would delay chemisorption reaction mechanism of less than 3%. Considering this result, we can easily deduce that native zirconia layer has no protective effect.

4.   The origin of SCC cracking is not chemical but mechanical. This point has been previously discussed. The SCC scenario would be able to predict SCC chemical risk without this assumption.

5.   The presence of oxygen localizes the phenomenon of SCC. As the numerical component is 1D axisymmetric, we cannot give any comment on this observation.

6.   The oxygen can lead to a passivation of the metal if the quantity of oxygen is important enough to induce formation of an oxide layer faster than the iodine attack. The proportion of oxygen is always higher in the gap compared to iodine, favoring the zirconia formation kinetic [Fig. 3]. The final balance is also in favor of zirconia formation [Fig. 5]. The theory underlying the scenario of thermochemistry confirms this observation.

Finally, the current scenario confirms five experimental observations over six. The sixth is not contradicted today, but not accessible with our 1D axisymmetric calculations.

7   CONCLUSIONS AND PERSPECTIVES

Thanks to the numerical component NUAGES, a sensitivity analysis has been performed allowing to confirm the protective effect of the zirconia layer against SCC.

- This protective effect looks to be not mechanical or thermal but only chemical.
- It can be quantified as follow:

$$Log\left(^t/_\tau\right) = Log\left(^t/_\tau\right)_0 + 0.167 e_{ZrO_2} - 0.044$$

with $Log\left(^t/_\tau\right)_0$ standing for $Log\left(^t/_\tau\right)$ in the case of a zero thickness zirconia layer.

e_{ZrO_2} represents the zirconia layer thickness [μm] and τ=1sec.

After coupling the zirconia layer model presented here and the SCC standard model, all hypotheses underlying the industrial modeling scenario remains valid, even strengthened.

The current modeling scenario agrees with five experimental observations over six. The sixth is not contradicted, but simply inaccessible today in our 1D axisymmetric approach.

An interesting perspective would be to enrich the present study with other simulations, in order to:

- Confirm the generic character of an equation to represent the impact of the zirconia against the SCC risk
- Refine when necessary the parameters of the equation modelling the delayed effect of zirconia on chemisorption kinetic.

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