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Comparison Between In-reactor Oxidation Behavior of Iron and Zirconium Alloys

Caio Melo[1] and Régis Reis[2]

[1]*Amazônia Azul Tecnologias de Defesa S.A. (Amazul)*
Av. Corifeu de Azevedo Marques, 1847
CEP: 05581-001 – São Paulo, SP, Brazil
caio.melo@marinha.mil.br

[2]*Instituto de Pesquisas Energéticas e Nucleares (IPEN)*
Av. Prof. Lineu Prestes, 2242
CEP: 05508-000 – São Paulo, SP, Brazil
regis.reis@ipen.br

1. Introduction

A number of early water-cooled power reactors utilized fuel rods clad with stainless steel. Its excellent thermomechanical properties and corrosion resistance permitted almost flawless operation of reactors of the PWR type [1]. In the early 1960s, a change to zirconium alloys took place largely motivated by economic reasons: the small thermal-neutron capture cross section of zirconium allowed reduced uranium enrichment, and generally improved fuel cycle efficiency. Adoption by the nuclear industry was relatively rapid, with the last water-cooled stainless steel cores operating no later than the 1980s.

The economically based choice of zirconium alloys, however, has been periodically questioned out of safety concerns, specially following nuclear incidents. Already in the 1970s, considerations of postulated accidents and TMI-2 brought attention to abnormal conditions phenomena such as clad oxidation, metal-steam reaction and hydrogen generation–conditions under which stainless steel might provide better safety margins [2]. The Fukushima nuclear accident in 2011 was a definitive example of the risks of hydrogen explosions following a loss of coolant situation, and once more put into question the use of zirconium in reactor design.

This work is intended as a contribution to the ongoing worldwide evaluation of new accident-tolerant clad materials [3]. More specifically, we continue the reassessment of stainless steel as clad initiated in [4] by introducing relevant corrosion models into a fuel rod simulation computer code and running comparative cases with other materials.

2. Methodology

In order to compare the performance of stainless steel clad fuel rods in actual reactor operation conditions, we employ TRANSURANUS [5], a computer program to simulate and permit the analysis of a variety of fuel rod designs. The code version employed as a starting point and which contains models for thermomechanical properties of austenitic stainless steel AISI 348 is that described in [4]. In that work, correlations developed with open literature data were introduced into basic TRANSURANUS for the following material properties: elasticity modulus, Poisson's ratio, irradiation-induced swelling, thermal strain, thermal conductivity, creep rate, yield stress, rupture strain, burst stress, specific heat, density and melting temperature. The modified code was validated against actual power plant data, showing reasonable agreement.

After modifying basic TRANSURANUS according to [4], low-temperature uniform corrosion and high-temperature oxidation models for stainless steel were gathered from published literature and introduced into the code.

The low-temperature uniform corrosion model was developed from [6], a long-term corrosion kinetics study of steel AISI 348 in ultra pure deoxygenated water and under relatively normal reactor operation conditions (360ºC and 20 MPa). The kinetic parabolic parameter for weight gain k_p is given by equation (1):

$$k_p = 0.135 \pm 0.014 \quad \left(\mathrm{mg/dm^2}\right)^2 \mathrm{d}^{-1}. \tag{1}$$

The expression for k_p in high-temperature oxidation conditions, given by equation (2), was obtained from [7], which investigates the behavior of AISI 304L and AISI 348 stainless steels in high-temperature (1000 ºC – 1350 ºC) steam environment:

$$k_p = 4.85 \cdot 10^7 \exp\left(\frac{-41338 \pm 1257}{T}\right) t \quad \left(\mathrm{kg/m^2}\right)^2 s^{-1}, \tag{2}$$

where T is in kelvin and t is in seconds.

Finally, the thermal conductivity for chromite, which represents the bulk of stainless steel oxides, was set as 3.0 W/m/K following [8].

Two cases have been selected for the assessment of stainless steel performance. A simpler, burnup accumulation only, hypothetical case freely provided by the European Commission's Joint Research Centre, responsible for the development of TRANSURANUS [9], and a more complete case, also burnup accumulation only, that simulates a fuel rod irradiated in Gravelines PWR. This latter case was modeled after data found in the freely available report [10].

3. Results and Discussion

Table 1 presents the peak oxide layer thickness values in micrometers (µm) for the two cases described above and four types of fuel rod clad material: the zirconium alloys Zircaloy-4 and M5, and the iron alloys FeCrAl and stainless steel AISI 348. With the exception of the last one, all the other materials are already modeled by the basic TRANSURANUS code version.

Table 1: Peak oxide layer thickness [μm] calculated with TRANSURANUS code

Material	**European Commission** *Burnup: 89.4 GWd/tU* *Coolant Temperature: 313 ºC*	**Gravelines** *Burnup: 76.6 GWd/tU* *Coolant Temperature: 348 ºC*
Zircaloy-4	44.1	38.4
M5	42.3	38.1
FeCrAl	6.8	6.0
AISI 348	1.4	1.2

From a qualitative point of view, the results seem reasonable. Zircaloy-4 exhibits larger corrosion than M5, and M5, in turn, a larger corrosion than FeCrAl. Since models for these materials have already been validated before inclusion into basic TRANSURANUS, the correct order of these values suggest that the two cases have been modeled consistently (geometrically, physically and with regards to boundary conditions). Hence, the much lower values for stainless steel may serve as evidence of its superior corrosion resistance, at least in a non-borated low-oxygen environment (we note once again the conditions under which the low-temperature corrosion data were obtained in [6]).

4. Conclusions

This work continues the reassessment of stainless steel as nuclear fuel rod clad in new reactor designs. Computer code simulations have shown evidence of superior corrosion resistance of steel in a normal operation setting, even in comparison with other iron based alloy.

Further research is still needed to show the feasibility of an industry-wide design change from zirconium alloys to stainless steel clad. Additional cases must be studied to confirm the lower steel values obtained earlier–most importantly, cases which exhibit abnormal (accident) conditions. Moreover, even for normal operation cases, the corrosion models must be corrected and extended to a variety of water chemical profiles.

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