

ADAPTATION OF FUEL CODE FOR LIGHT WATER REACTOR WITH AUSTENITIC STEEL ROD CLADDING

Daniel de Souza Gomes¹, Antonio Teixeira e Silva¹ and Claudia Giovedi²

¹Instituto de Pesquisas Energéticas e Nucleares (IPEN / CNEN - SP)
Av. Professor Lineu Prestes 2242
05508-000 São Paulo, SP
dsgomes@ipen.br, Teixeira@ipen.br

²Laboratório de Análise, Avaliação e Gerenciamento de Risco (POLI / USP - SP)
Av. Prof. Mello Moraes 2231
05508-000 São Paulo, SP, Brazil
claudia.giovedi@labrisco.usp.br

ABSTRACT

Light water reactors were used with steel as nuclear fuel cladding from 1960 to 1980. The high performance proved that the use of low-carbon alloys could substitute the current zirconium alloys. Stainless steel is an alternative that can be used as cladding. The zirconium alloys replaced the steel. However, significant experiences in-pile occurred, in commercial units such as Haddam Neck, Indian Point, and Yankee experiences. Stainless Steel Types 347 and 348 can be used as cladding. An advantage of using Stainless Steel was evident in Fukushima when a large number of hydrogens was produced at high temperatures. The steel cladding does not eliminate the problem of accumulating free hydrogen, which can lead to a risk of explosion. In a boiling water reactor, environments easily exist for the attack of intergranular corrosion. The Stainless Steel alloys, Types 321, 347, and 348, are stabilized against attack by the addition of titanium, niobium, or tantalum. The steel Type 348 is composed of niobium, tantalum, and cobalt. Titanium preserves type 321, and niobium additions stabilize type 347. In recent years, research has increased on studying the effects of irradiation by fast neutrons. The impact of radiation includes changes in flow rate limits, deformation, and ductility. The irradiation can convert crystalline lattices into an amorphous structure. New proposals are emerging that suggest using a silicon carbide-based fuel rod cladding or iron-chromium-aluminum alloys. These materials can substitute the classic zirconium alloys. Once the steel Type 348 was chosen, the thermal and mechanical properties were coded in a library of functions. The fuel performance codes contain all features. A comparative analysis of the steel and zirconium alloys was made. The results demonstrate that the austenitic steel alloys are the viable candidates for substituting the zirconium alloys.

1. INTRODUCTION

During the 1970s, commercial units worked with Stainless Steels (SS) as fuel cladding. The alloys utilized during the 1960-1975 period included SS Types 304, 304L, 316, 347, and 348 [1]. In this epoch, power reactors such as Haddam Neck, Indian Point 1, Yankee Rowe, and Lacrosse were operated with the SS rod cladding, showing an excellent conduct. The steel cladding was heat treated, annealed, and cold-worked.

The good performance record of austenitic steel clad fuel used in both stations Yankee Rowe, and San Onofre 1 plant between the years from 1970 to 1980 was forgotten. The LaCrosse reactor reported cladding failures in fuel pins when using SS Type 304. The cause was determined to be intergranular cracking after only 500-1000 h of operation [2]. Therefore, zirconium base alloys replaced steel as the cladding material beginning in the early 1970s as documented by Fukushima and Chernobyl. The zirconium alloy cladding provided improvements because the heat treatment applied reduced the number of failures. Currently, austenitic stainless steels are extensively used as structural material in nuclear applications, and the Types 304 and 316 are used in control rods as cladding [3].

The overall aims of this study include an exam of the properties of Types 321, 347 and 348. In stainless steel, the elements as niobium, titanium, and tantalum can occur. Tantalum content is ten times the minimum carbon content. The steel Type 321 is similar to Type 304 except for the addition of titanium, that helps prevent chromium carbide precipitation. The investigation involved thermal and mechanical characterization of the stabilize alloys. The objective was the creation of a modern code that could use the Type 348 as fuel cladding. A new library produced to contain the functionality to calculate the cladding performance using SS type 348.

The new fuel code contains the steel properties that were researched and compiled in FORTRAN planned to substitute the equivalent correlation and properties of zirconium alloys. The main purpose of the investigation was to create a system that could simulate the fuel behavior for an extended irradiation cycle such as steel cladding. The solution adopted was the codification of an original library of 348 steel. The task involved the definition of the thermal and physical properties to build the code so that the system could simulate the stable state. However, the reactors of the epoch using steel cladding had low burnup ranging from 20 to 30 MWd/kgU.

2. BACKGROUND

The fuel code applied to this study was the Thermal-Mechanical Behavior of Oxide Fuel Rods (FRAPCON) sponsored by the North American Nuclear Regulatory Commission. FRAPCON is a licensed code intended for use in commercial nuclear plants. Advanced alloys may meet the best conditions for extended irradiation cycles at 18 to 24 months, or burnup over 62 MWd/kgU. An accident tolerant fuel (ATF) must show higher resistance to neutron flux. The ATF program proposes a rational choice of alloys involving new materials. The candidates found by ATF program are metallic and ceramic, such as stainless steel with higher chromium, or silicon carbide [4].

2.1. A Comparison Between Zirconium Alloys and Austenitic Steel

Pressure vessels and structural materials continue to be made of steel. Zirconium alloys could also be used in the permanent parts of commercial reactors. Zircaloy cladding provides better neutron utilization than steel because of the reduced cross section. During the 1990s, steel was abandoned fully because of the drop in the price of zirconium. Furthermore, the properties exhibited by zirconium alloys must support the necessary mechanical properties for building 3.6-m long tubes for storing uranium dioxide [5].

The Yankee unit was used steel cladding, in a pressurized water reactor (PWR). The Yankee net power was 134 MWe with fuel enriched to 2.6%. The third-generation reactors operated with the fuel enriched up to 4% used zirconium alloys and U-235 because of the smaller cross-sections than steel. The process of replacing steel with zirconium-based alloys, instead of less-expensive SS, took about two decades. Around 1960, the investigations begun about of steel behavior under radiation. The properties of Type 347 and 348 were measured in the fast breeder reactor (FBR).

The strain fatigue experienced while annealing these types of steel increases the yield strength, varying in the degree of cold working. The phases of zirconium alloys transform the crystal from body-centered cubic (bcc) to hexagonal close packed (hcp) at 830 °C. Stainless steels have a face-centered-cubic (fcc) structure. Table 1 describes the properties of austenitic steels and Zircaloy-4 [6].

Table 1: Properties of Zircaloy-4 and steel Types SS-321, SS-347 and SS-348

Properties	Zircaloy-4	SS-347	SS-348	SS-321
Crystal structure	hcp(α)/bcc(β)	fcc	fcc	fcc
Density 10^3 .(Kg/m ³)	6.56	7.840	7.840	7.840
Hardness Rockwell-B	89	85	85	85
Tensile strength, ultimate (MPa)	413	620	655	620
Tensile strength, yield (MPa)	241	240	275	240
Elongation at break (%)	20	50	45	45
Modulus of elasticity (GPa)	99.3	195	195	193
Poisson ratio	0.37	0.27	0.27	0.27
Shear modulus (GPa)	36.2	77	77	77
Electrical resistivity μ (Ohm-cm)	74	73	79	72
CTE, linear ($\mu\text{m}/\text{m}\cdot^\circ\text{C}$)	6	17.3	17.3	16.7
Specific heat capacity (J/g $\cdot^\circ\text{C}$)	0.285	0.5	0.5	0.5
Thermal conductivity (W/m $\cdot^\circ\text{C}$)	21.5	16.3	16.3	16.1
Melting point ($^\circ\text{C}$)	1850	1400	1400	1400
Cross section (barns)	0.184 σ_c	3.03 σ_c	3.13 σ_c	3.06 σ_c

2.2. Intergranular Stress Corrosion Cracking

The low-carbon or austenitic steel is a type of chrome-nickel alloy, non-magnetizable. Steel Types 347 and 348 use chemical stabilizers as niobium. The alloys referenced were resistant to carbide precipitation. Therefore, planned to work within the temperature range, where carbide precipitation develops. The hydrogen promotes the grain boundary embrittlement, following the cracking. The hydrogen diffusion may accelerate the cracking process [7]. During the corrosion occurs higher exposure will lead to an increasing susceptibility to cracking [8]. The hydrogen produces intergranular stress corrosion cracking (IGSCC) in steel and zirconium alloys. It observed an attractive decrease of IGSCC, in the Types 347, 348. The low corrosion rate also occurs in Type 321.

Both the Types 347 and 348, contain niobium and tantalum. Irradiation must follow hydrogen diffusion to form lattice structure becoming brittle, adding hydrogen, and reducing the carbides as $(\text{Fe, Cr, Ni, Mo})_{23}\text{C}_6$.

2.3. Hydrogen Generation During Nuclear Accidents

The hydrogen generation occurs during nuclear accidents. In disaster exist evidence of a meltdown at Three-Mile Island (TMI), in March 1979. A comparison between Chernobyl, April 1986, and Fukushima, March 2011, showed to dominating evidence of apparent disadvantage of using the zirconium alloys [9]. The TMI accident became a turning point in the safety design of nuclear reactors. The steels suffered damages because of intergranular attack, in boiling water reactor (BWR). Steel alloys perform better in PWRs. The control rods can manage the reactivity, using cladding made of stainless steel as types 304 or 316. The control rods were formed by a range of 20 to 89 rods and pressurized by helium.

2.4. Accident Tolerance Fuels

The research supports the use of coatings such as silicon carbide (SiC) or iron-based alloys. The alloy could be iron-chromium-aluminum, preferred in the ATF program [10]. Most likely, the monolithic ceramic fuel will replace the UO_2 . Future designs must improve the fuel performance during transients, design-basis events, and beyond design-basis events. Furthermore, it must show geometric stability, low ductility reduction, and enough resistance to oxidation. The rod coated with SiC shows lesser damage at elevated temperatures. The new pellet must have a higher density by including materials like uranium silicide (U_3Si_2).

2.5. Fuel for Supercritical Light Water Reactor

The fourth-generation supercritical water reactor (SCWR) promises an improvement of thermal efficiency from 35% to 45% [11]. The fuel planned for the SCWR is a ceramic pellet consisting of less than 5% of enriched uranium. The pellet can also use plutonium, thorium, or mixed oxides. The typical operating temperature will be over 600 °C. The materials selected for fuel cladding should have good ductility and high corrosion resistance. The austenitic SS alloys with elevated chromium concentration, such as steel Types 304H or 310H, are candidates. The chromium concentration improves oxidation resistance when the level reaches between 18 and 26. For an SCWR, steel with 25% chromium concentration is one of the best choices for cladding.

3. MATERIAL AND METHODS

In this investigation, the capability to predict the behavior of the steel cladding is demonstrated. A fuel code helps in determining the average burnups during cladding with SS. The burnup cycle in 1970 was from 20 to 30 MWd/kgU. The results compare the difference between the fuel cladding made of zirconium alloys, and SS Type 348. The pellet and cladding mechanical interaction is probably the major contributor to fuel element failure. Therefore, it is important to understand when hard pellet-clad contact takes place. The fuel initially densifies and begins to swell after 10 MWd/kgU. The contact occurs over the region of swelling even though the fuel appears to be densifying. In reality, the fuel suffers a mechanical contraction due to gap closure and improves the thermal conductivity.

3.1. US-PWR 16x16 LTA Extended Burnup Demonstration Program

The objective of the investigational program was to examine the fuel management efficiency under an extended discharge burnup with a maximum of 58 MWd/kgU. The program was conducted during the 1980s. The effects created by fission products, and the degradation of structure materials must also be researched. Table 2 displays the thermal hydraulic parameters of the fuel rod TSQ002.

Table 2: Thermal-hydraulic parameters

Thermal-Hydraulic	Values
Thermal power (MWt)	2815
Heat generate in fuel(%)	97.4%
Pitch (mm)	12.7
Nominal pressure (MPa)	15.51
Minimal pressure on steady state (MPa)	16.17
Core inlet temperature (°C)	289.4
Core outlet temperature(°C)	322.5
Average linear power density (kW/m)	17.75
Maximum linear power density(kW/m)	41.67
Core coolant flow rate (kg/h)	54.61x10 ⁶
Average speed along the fuel rod (m/s)	4.99

The fuel rod irradiated in the US-PWR 16 x 16 reactors used Zircaloy-4 as cladding. The fuel assembly design was formed by 236 rods with five control elements and 12 spacer grids. This fuel rod was used in the International Fuel Performance Experiments. In this case, the fuel rod cladding was originally Zircaloy-4. The full-length rods were irradiated to an assembly, and lead rod average discharge cycles ranged from 52 to 58 MWd/kgU. Table 3 displays the input parameters to performance fuel code of the experimental fuel rod TSQ002.

Table 3: Input parameters for performance fuel code

Input Parameters	Values
Irradiation time (days)	1671
Neutron flux (n/m), (E>0,82MeV)	5.41x10 ¹²
Neutron flux thermal (n/m)	26 x10 ¹⁵
Plenum length (m)	0.2717
Inner pressure of gas helium (MPa)	2.62
Water pressure (MPa)	15.5
Cooling mass flux (Kgs/m ²)	5899.60

The alloys used to fabricate the separator grids were made of nickel-chromium-molybdenum. Niobium and tantalum were added to Inconel 625, which stabilized and hardened the matrix. A spring forced the pellet stack to stay within the cladding.

The spring was located in the plenum, holding the fuel column in position. The test consists of assembling the input file, which calculates the output parameters during 1671 days of power generation. The standardized designs of the fuel rod include ceramic fuel and enriched uranium dioxide at 3.48%. In the simulation, 70 periods were used to represent the days of irradiation. The important point is a corrosion mechanism that is producing ZrO_2 in the zirconium alloys, but the steel produce irons oxides. The behavior of oxidized alloys is different. The input parameters used for performance code, for the rod TSQ002, is shown in Table 4.

Table 4: Fuel and cladding parameters of experimental fuel rod

Fuel and Cladding	Values
Cladding outer diameter(mm)	9.70
Cladding inner diameter (mm)	9.50
Cladding thickness (mm)	0.635
Gap thickness (mm)	0.088
Fuel high (mm)	0.90
Fuel outer diameter UO_2 (mm)	8.22
Fuel enrichment (%)	3.48
Fuel dish (mm)	0.34
Pellet density (g/cm^3)	10.60

4. DISCUSSION AND RESULTS

After included correlations and coded the properties of steel 348, executed the simulation. The new system is composed of about twenty fundamental routines. The central fuel temperature depends on the generated power showing the same format, as in Figure 1.

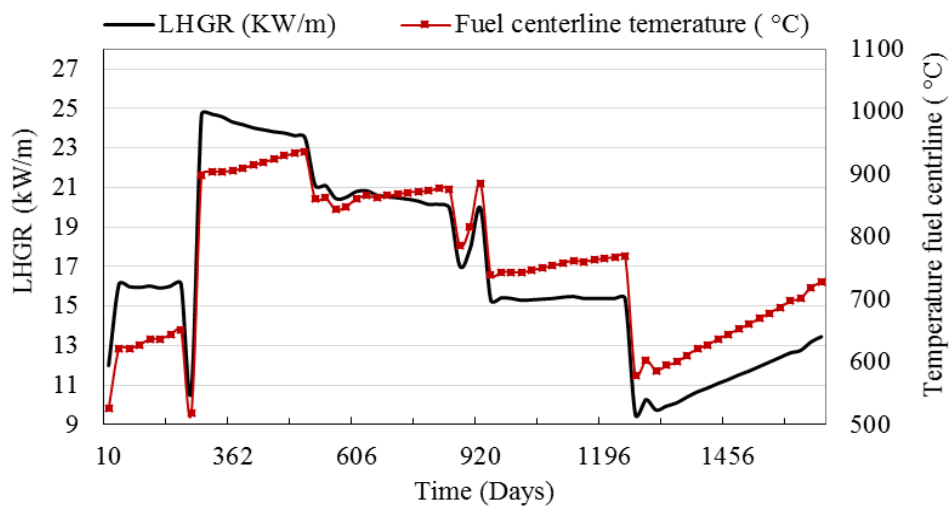


Figure 1: Linear power rate and centerline pellet temperature steel 348

The data were simulated with a new library that included the correlation of steel Type 348 properties. In the simulation of US-PWR 16 x 16, the fuel produced higher temperatures than when Zircaloy is used, because of an elevated heat transfer rate of the steel. The results showed that the gasses generated by fission amounted to 0.24% with Zircaloy, and 0.64% with steel, because of the elevated temperatures. Figure 2 displays the outside deformation of both claddings.

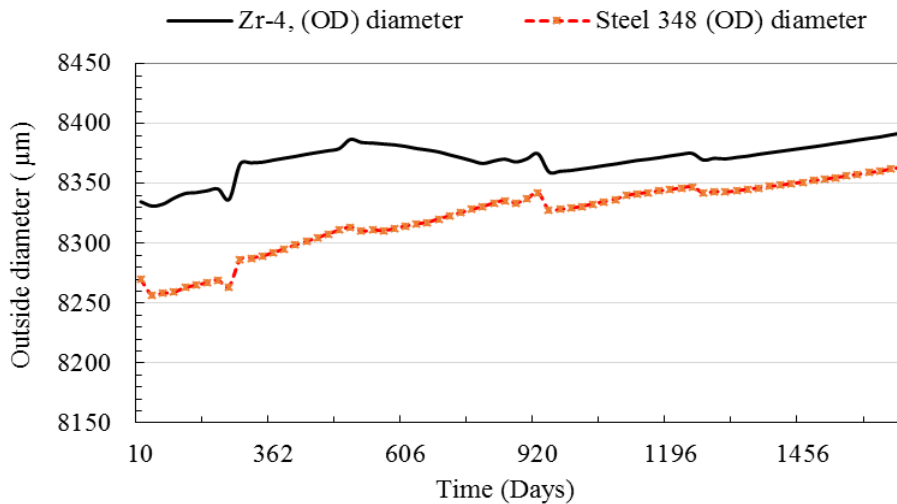


Figure 2: Outside diameter contrast between Zr-4 and steel 348.

The elastic modulus of steel Type 348 reaches 195 GPa, whereas the young modulus of Zircaloy-4 is 99.3 GPa at room temperature. During the irradiation cycle, a lower deformation should occur in the steel even though other factors are acting dynamically. Radial distortion in the Zircaloy-4 was higher by 0.57%. Figure 3 shows that the axial deformation in steel was greater than in Zircaloy-4. The differences are caused by differing heat transfer properties of the materials and the thermal expansion in a 3.8 m tube.

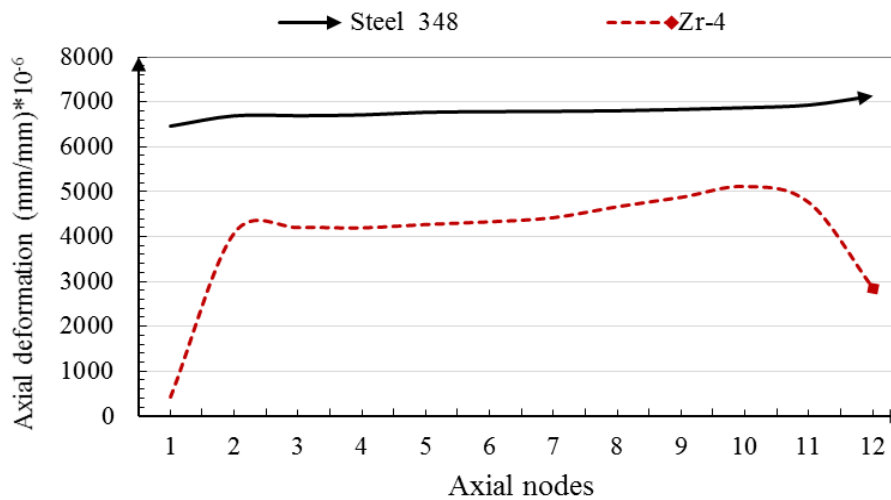


Figure 3: Axial deformation contrast between Zr-4 and steel 348.

Fuel heating occurs because the conductivity of Zircaloy exceeds that of steel by 30%. The thermal conductivity of Zircaloy is 21.3 W/mK and 16.3 W/mK for steel. Significant thermal effects are caused because the specific heat of steel is 0.5 J/Kg°C and 0.28 J/Kg°C for Zircaloy, and fuel fission generates the same amount of heat. The temperatures are similar in both cladding types, as plotted in Figure 4. The differences are due to the differences in thermal conductivity and heat capacity of materials.

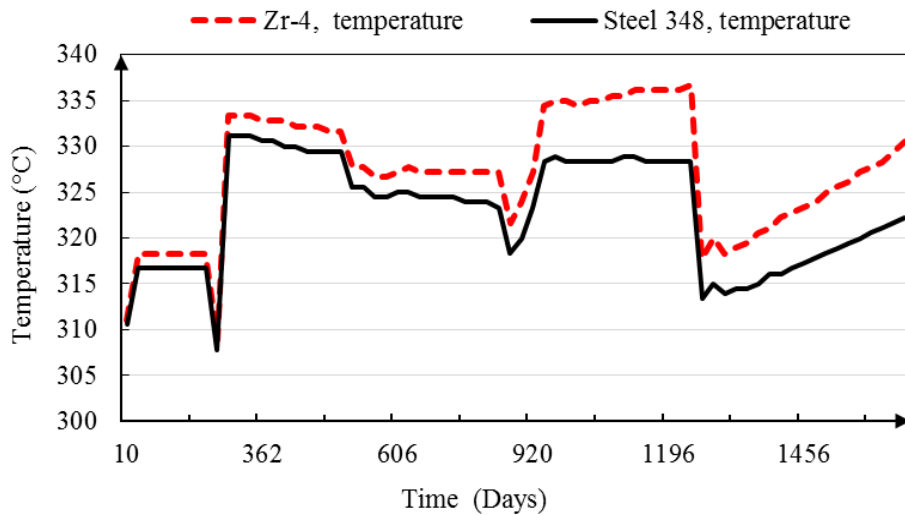


Figure 4: Temperature between Zr-4 and steel-348.

The temperatures in the cladding are similar to the zirconium alloys and steel. The gap closure is one of the factors defined as crucial to reducing fuel degradation. In the case of steel cladding, the superiority of Type 348 is clear; however, Zircaloy-4 outperforms Type 348. The Zircaloy-4 closes at 553 days of irradiation, but the stainless steel closes the gap after 1100 days. Following the closure, the contact between the pellet and the inner wall of cladding occurs and the failure risk increased. The gap closure behavior is similar, but Zircaloy achieves contact before the steel, as shown in Figure 5.

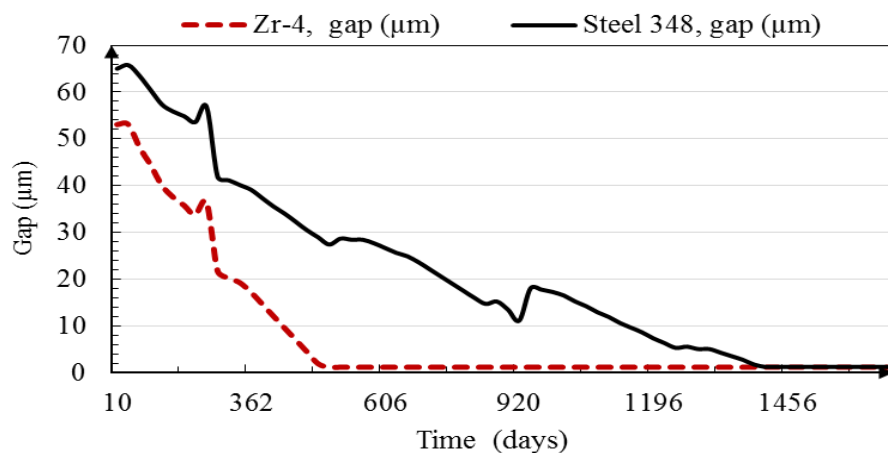


Figure 5: Gap closure between Zr-4 and steel 348.

The predicted creep rate is a function of the applied stress. The irradiating flux is independent of temperature for SS. The mechanical behaviors of austenitic steels and Zircaloy-4 are significantly different. The temporal response, produced by operational loads, was considered to be slower in Type 348. However, because of reductions in the area and permitting local deformations, the stresses are as shown in Figure 6.

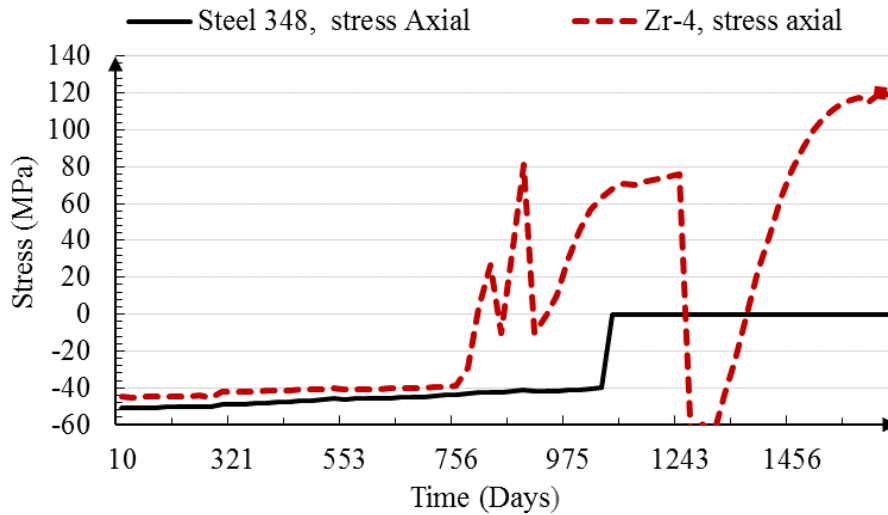


Figure 6: Stress produced by the difference between Zr-4 and steel 348.

Measuring the effects of diffusion on hydrogen and oxygen for the steel is a complicated and theoretical field that is left for a future research. The exposure level is the main impact of radiation in the fuel rod cladding. The increased mechanical strength and decreased uniform elongation of the alloys also affect cladding. Besides, embrittlement due to hydrogen diffusion and oxidation at high temperatures can rupture the coating.

4.1. Effects of Irradiation Damage

The steel Type 348 demonstrates the best features to substitute the zirconium alloys when compared with Type 347 or Type 321, but with similar performance. The modulus of elasticity of steel is higher and more stable about corrosion. The prime gamma elements, such as nickel, stabilize against martensitic transformation. The addition of molybdenum, phosphorus, and silicon improves the quality of steel Type 348. The steel Type 348 supports stronger deformations (up to 3.0%) than commercial steels (0.2% to 1.0%) when irradiated. The thermal expansion at room temperature of steel is greater when compared with zirconium alloys. The elongation of steel is higher than zirconium by approximately 20%.

The thermal conductivity of steel is 75% of the conductivity of Zircaloy. In the SS cladding, heat flows more slowly, producing heat-up of fuel. Therefore, SS must generate more fission gasses due to the increase of inner pellet temperature. An analysis of the mechanical modules without irradiation shows reductions caused by crystal damages. The elevated temperatures produce a nominal value inferior for elastic moduli. The results of the test confirm that the reactor could use steel.

Type 348, according to the data provided by the code, with adjustments. The results of the analysis take into account the behavioral complexity of steel in irradiation conditions.

5. CONCLUSIONS

The library implemented for the austenitic stainless steel Type 348 can predict the fuel performance. The fuel rod TSQ002 is a basis for the results obtained. The outcomes are consistent with the outcome of the reactors coated with steel. The SS can be used as cladding in five power reactors. The stainless steel offers advantages because of small mechanical weakening and the presence of a few hydrides.

However, steels present a high neutron cross section. The Types 347 and 348 are more resistant due to stabilization than Type 321, with a better creep rate under irradiation. The stabilized steels must produce a lower density carbide around of the grain. The IGSCC process must develop chromium oxide film with 0.02- μm thickness.

ACKNOWLEDGMENTS

The authors are thankful for the support received from Nuclear Energy Research Institute (Instituto de Pesquisas Energéticas e Nucleares; IPEN). Associated the National Nuclear Energy Commission (Comissão Nacional de Energia Nuclear; CNEN).

REFERENCES

1. J.M. Beeston, "Mechanical and physical properties of irradiated type 348 stainless steel." In: Effects of Radiation on Materials, *Proceedings of the Eleventh International Symposium: a Symposium. ASTM International*, pp. 303.(1982).
2. A.Strasser, J. Santucci, K. W. Yario, G. Stern, L. Goldstein, L. Joseph, "An Evaluation of Stainless Steel Cladding for Use in Current Design LWRs, NP-2642". *Electric Power Research Institute, Palo Alto, CA*, pp. 251 (1982).
3. D. L. Hagraman, G. A. Reymann, "MATPRO-VERSION 11: a handbook of materials properties for use in the analysis of light water reactor fuel rod behavior". *Idaho National Engineering Lab., Idaho Falls (USA)*, pp. 681 (1979).
4. K. L. Geelhood et al. "FRAPCON-3.4: A Computer Code for the Calculation of Steady State Thermal-mechanical Behavior of Oxide Fuel Rods for High Burnup". *Richland, WA: US Nuclear Regulatory Commission, Office of Nuclear Regulatory Research* (2011).
5. K. A.Terrani, S. J. Zinkle, L. L. Snead, "Advanced oxidation-resistant iron-based alloys for LWR fuel cladding." *Journal of Nuclear Materials*, Volume **448**, n. 1, pp. 420-435 (2014).
6. D. Peckner, I. M. Bernstein, *Handbook of Stainless Steels*, New York, McGraw-Hill Book Company (1977).
7. A. G. Varias, A, R. Massih, "Simulation of hydrogen embrittlement in zirconium alloys under stress and temperature gradients.", *Journal of Nuclear Materials*, Volume. **279**, n. 2, pp. 273-285, (2000).

8. F. A. Garner et al. "Assessment of XM19 as a Substitute for AISI 348 in ATR Service". INL/EXT-07-13530, *Idaho National Laboratory, November 2007*. Number of cycles to failure 103 10-1 100 2 5 2 5 EC-316L: 430 C Nonirrad. Irrad.: 10 dpa Plastic strain range (2007).
9. A. Abe, C. Giovedi, D. S. Gomes, A. T. Silva, "Revisiting Stainless Steel as PWR Fuel Rod Cladding after Fukushima Daiichi Accident". *Journal of Energy and Power Engineering*, Volume. **8**, n. 6 (2014).
10. K. G. Field, "Irradiation Stability of ATF FeCrAl". ADVANCED FUELS CAMPAIGN FY 2014 ACCOMPLISHMENTS *REPORT* Prepared for the U.S. Department of Energy Office of Nuclear Energy Under DOE Idaho Operations Office Contract DE-AC07-05ID14517, (2014).
11. J. Buongiorno, P. Macdonald, "Supercritical water reactor (SCWR)". *Progress Report for the FY-03 Generation-IV R&D Activities for the Development of the SCWR in the US*, INEEL/Ext-03-03-01210, INEEL, USA, (2003).