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SIMULATION OF NUCLEAR FUEL EXPERIMENTS USING DIONISIO FEM

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Abstract: DIONISIO FEM is a new module incorporated into the DIONISIO code, developed by the Codes and Models Section of the Nuclear Fuel Cycle Management, CNEA. This module uses the core of the fuel code, including the mesh generator, the materials library, the solvers of equations systems and finite element libraries, to simulate the pellets behavior subjected to different thermal conditions. Based on this framework, the FEM module solves a thermomechanical problem, in the required geometry, depending on the material and the physical and / or chemical models to be evaluated. The results obtained for the dimensional evolution suffered by a fuel pellet when it is subjected to a purely thermal history are presented. In general, simulations carried out with DIONISIO FEM achieved good agreement with the experimental data.

1 INTRODUCTION

DIONISIO is a nuclear fuel code that simulates the main phenomena that occur within a fuel rod during irradiation in a nuclear reactor under normal or accident conditions. The code is organized in a modular structure with several interconnected models. Among the predictions of the code are: the thermochemical and thermomechanical evolution of a nuclear fuel rod, the fission gas release process, the thermohydraulic behavior of the coolant surrounding the cladding and the pellet-cladding mechanical interaction (PCMI) (Denis and Soba, 2003; Soba, 2007; Soba and Denis, 2008, 2015). To solve the pellet-gap-cladding system, the Finite Element Method (FEM) is implemented in a two- or three-dimensional domain, at the user's choice. In the former case, cylindrical symmetry has to be assumed. In addition, subroutines to simulate the fuel behavior in high burnup conditions, as well as LOCA type conditions have been incorporated within the code (Lemes et al., 2015, 2017, 2019; Soba et al., 2013, 2014). Moreover, the models and laws used are in permanent review in order to improve the predictions' reliability (Cazado et al., 2021; Cazado and Denis, 2018; Goldberg et al., 2019).

All the descriptions given above correspond to simulations of fuel rods under irradiation. Nevertheless, different experiments of materials for the nuclear industry take place in out-of-pile conditions. In these experiments, the material is subjected to a specific thermal or pressurization history or it can be tested to determine its mechanical properties. In some cases, the samples can be expose to steam, hydrogen, or high concentration of iodine to study their influence on the material under testing (Erbacher, 1981; Grieger et al., 1999; Hózer et al., 2001, 2005; Karwat, 1985; Markiewics and Erbacher, 1988; Perez-Feró et al., 2010; Powers and Meyer, 1979; Vasáros and Matus, 1999). Likewise, several out-of-pile experiments evaluate the effects of temperature, atmosphere and microstructure on fuel pellets during well-controlled processes (Brite et al., 1975; Cunningham et al., 1981; Hann et al., 1977).

Based on the finite element method, a new module named DIONISIO FEM was created to analyze the experiment described above. This module uses the DIONISIO code kernel including finite element and materials libraries, equations system solvers and mesh generator.

In this paper, attention is paid specially to densification experiments involving UO_2 pellets while the simulations of the cladding tests will be presented elsewhere.

A densification model, briefly described here, was developed to be used on irradiated samples as well as for thermal pellet characterizations. The densification model elaborated was tested by comparing its predictions with experimental data published in the open literature. The first simulations, that were published elsewhere (Cazado, 2019; Cazado and Denis, 2018) were aimed at testing the model separately, and hence the equations involved were solved under certain simplifying assumptions (considering grains of average size and isothermal conditions). The influence of temperature and initial pores size distribution on the densification rate was analyzed. Although a good agreement between the calculated and measured valued was obtained, the need of evaluating the dimensional changes with the local conditions within the pellet became evident. Hence, the densification model was incorporated to the DIONISIO code. It is worth mentioning that a tool like DIONISIO FEM is particularly useful to predict the irradiation behavior of a material which initial microstructure is not very well known. With the experimental results of the fuel re-sintering tests, which are usually provided, different pore distributions can be proposed and the simulated case that meets the thermal test conditions can be selected as the most suitable estimate to run the code for experiments under irradiation conditions.

This paper is organized as follows: in section 2, the porosity model previously developed is briefly described. In section 3, several results corresponding to experiments without irradiation are presented. In the last section some conclusions are given.

2 MODEL DESCRIPTION

The dimensional changes of UO_2 fuel pellets under irradiation conditions are of important concern due to possible pellet-cladding mechanical interaction that can impose constraints on a reactor operation. Densification, i. e., the reduction of as-fabricated pore sizes, is predominant during the first stages of fuel irradiation. Along with swelling, they are key in determining the physical changes that the fuel will suffer in a given power history (Assmann and Stehle, 1978; Maier et al., 1988; Olander, 1976). Generally, the fuel pellet behavior is characterized through experiments performed outside the reactor (out-of-pile studies) and without irradiation since in-reactor tests are complex and expensive (Basov, 2009). In a re-sintering test, a basic out-of-pile method, the dimensional stability of a fuel pellet is measured after subjecting it to a given thermal cycle in a controlled atmosphere. Fuel manufacturers uses the density change measured in these experiments as estimators of the in-reactor densification. Based on the diffusion of point defects, a model describing the evolution of the fuel pellets microstructure under thermal and irradiation conditions was previously presented (Cazado and Denis, 2018). The main considerations of the proposed densification model are described below:

- The fuel material is considered to be stoichiometric UO_2. In this sense, point defects evaluated here consist of one U cation and two O anions.
- Porosity in a UO_2 pellet may be located within the grain (intragranular pores) or between two grains (intergranular pores).
- Under purely thermal conditions, the concentration of point defects can be expected to take values close to those of thermodynamic equilibrium. Interstitial formation energy is higher than for vacancy, hence interstitial self-diffusion is negligible.
- Crystallographic defects include vacancies, edge dislocations, grain boundaries, pores.
- Dislocations climb occurs as a consequence of point defects.
- The pores decrease in size by releasing vacancies and expand on account of vacancies uptake.
- The rate of change of intergranular pores size is affected by the grain boundary self-diffusion while for intragranular pores only bulk self-diffusion is assumed.

The densification process described above is governed bulk and grain boundary self-diffusion of point defects in the UO_2 lattice. An extensive amount of experimental data on this matter can be found in the open literature (Alcock et al., 1966; Auskern and Belle, 1961; Matzke, 1987; Reimann and Lundy, 1969; Reynolds and Burton, 1979; Sabioni et al., 1998, 2000; Yajima et al., 1966). Nevertheless, the diffusion coefficient measures showed a considerable dispersion that may obey to different reasons: the methods used have non-comparable precision ranges, the samples presented different microstructures (single or poly-crystalline materials) or have unlike porosity degree or pores distribution (intra or inter-granular pores,), etc. A comparative study has been presented in Cazado (2019).

The diffusion coefficients considered in this work are models with:

$$D_U = 4.3 \times 10^{-8} \exp\left(-\frac{3.79[eV]}{kT}\right) \ [\mathrm{m}^2/\mathrm{s}] \tag{1}$$

$$D_{U_{GB}} = \begin{cases} 3.07 \times 10^{-6} \exp\left(-\dfrac{3.05[eV]}{kT}\right) & T < 1973\ K \\ 2.15 \times 10^{-2} \exp\left(-\dfrac{3.79[eV]}{kT}\right) & T \geq 1973\ K \end{cases} \quad [\mathrm{m}^2/\mathrm{s}] \tag{2}$$

where D_U and D_{UGB} are the bulk and grain boundary self-diffusion coefficients for UO_2, k is the Boltzmann constant and T is the absolute temperature.

Densification is quantified by evaluating the density change ($\Delta\delta$) of the fuel material, or the relative density change ($\Delta\delta/\delta_0$) or the relative volume change ($\Delta V/V_0$), the two latter referred, respectively, to the density or volume of the as-fabricated material, respectively.

3 RESULTS AND DISCUSSIONS

The model described in section 2 was subjected to an extensive examination program that started with a number of separate tests. These evaluations were aimed at optimizing the model parameters. Furthermore, they revealed the importance of possessing a correct description of the initial pore distribution. Some of these results were presented in a previous paper (Cazado and Denis, 2018) and consisted in comparisons between calculated and experimental data.

The present section presents the results obtained with the densification model in DIONISIO FEM.

Densification data reported by Maier et al. (1988) was simulated using the new module. Pellets corresponding to four different microstructures were manufactured by cold pressing and on a variety of sintering conditions. The re-sintering experiments consisted of a heating ramp at 1200 K/h up to 1973 K in a pure hydrogen atmosphere and held at this temperature during 280 h. Due to no details were given regarding the cooling rate, in this work we assumed it to be equal to the heating rate. The densities were measured using the xylene immersion technique. The general microstructure characteristics of these fuels are listed in Table 1.

Table 1 - Microstructural characteristics of the fuel pellets reported by Maier et al. (1988)

Fuel Type	Grain Size (µm)	Mean density (% TD)	Pore Structure	Mean Volume Pore Diameter (µm)
M-1	6.5	93.38 ± 0.20	Monomodal	2.0 - 3.0
M-2	9.5	94.70 ± 0.20	Bimodal	10.0
M-3	6.5	96.51 ± 0.20	Monomodal	2.0 - 3.0
M-4	8.5	96.05 ± 0.20	Monomodal	2.0 - 3.0

In Figure 1, a comparison between experimental porosity change data (Maier et al., 1988) and the results calculated with DIONISIO is presented. The time values given include the correction on account of the heating and cooling ramps during the tests. The relevance of incorporating heating and cooling ramps in the simulation was stablished in a previous work (Cazado, 2019), particularly for porosity distributions with a large population of small pores and for temperature above 1700 K. It can be appreciated that for most of the fuel types, the major porosity change occurs before 50 hours of treatment and since then, the porosity reduction rate decreases. Some deviations can be observed, especially for fuel type M-1, for which the experimental densification before 100 hours of treatment was lower than calculated while after that it was higher. This may be attributed to possible numerical misrepresentation of the experimental pore distribution skewness for that particular case. Despite this, there is a good agreement between experimental data and our results in most cases.

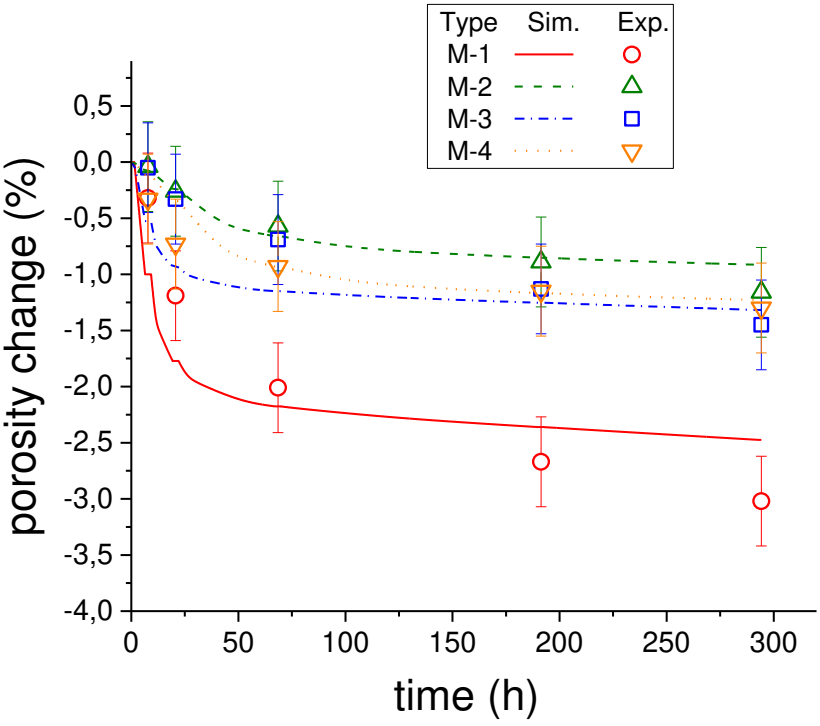

Figure 1 - Porosity evolution simulation for samples re-sintered at 1973 K up to 280 h. of isothermal treatment and its comparison with experimental data reported by Maier et al. (1988).

Experimental data corresponding to fuel pellets manufactured for the Instrumented Fuel Assembly (IFA) -431, -432 -527 and -513 (Cunningham et al., 1981; Hann et al., 1977) were simulated with DIONISIO FEM. The pellets were fabricated from 10% enriched UO_2 powder and sintered in a hydrogen and argon atmosphere. Nominal pellets diameter and length were 10.68 mm and 12.70 mm, respectively. Pellets with nominal densities of 92% TD and 95% TD with different pore distributions were included in these tests.

In Table 2 and Table 3 are listed the microstructural and geometric properties for different fuel type pellets used in IFA experiments, where d_p stands for pore diameter.

Table 2 - Microstructural characteristics of the fuel pellets used in IFA experiments (Cunningham et al., 1981; Hann et al., 1977)

IFA	Fuel Type	Mean Grain Size (μm)	Porosity (%)			Median Pore Diameter (μm)		
			d_p < 1 μm	d_p > 1 μm	d_p > 10 μm	d_p < 1 μm	d_p > 1 μm	d_p > 10 μm
431/432	IFA-1	6	3.1	4.9	2.4	0.6	8.5	32
431/432	IFA-2	10	1.6	6.4	5.9	0.3	36	36
431/432	IFA-3	56	0.4	4.6	1.3	0.6	5.1	32
513	IFA-4	12	0.4	4.6	2.1	0.7	7.3	28
527	IFA-5	12	0.6	4.4	1.8	0.7	5.6	36

The re-sintering tests for IFA 431 and 432 were performed under five different conditions combining temperature and isothermal time. Holding time for 1873 K was 24 hours and for 1973 K, 4, 7.8, 24 and 48 hours. The controlled atmosphere consisted of Ar-8%H_2 and cooling and heating rates reported by the authors were 300 K/h.

Table 3 - Geometric characteristics of the fuel pellets used in IFA experiments (Cunningham et al., 1981; Hann et al., 1977)

IFA	Fuel Type	Length (cm) ± 2σ	Diameter (cm) ± 2σ	Density (% TD) ± 2σ
431/432	IFA-1	1.296 ± 0.009	1.0679 ± 0.0001	92.10 ± 0.21
431/432	IFA-2	1.288 ± 0.006	1.0678 ± 0.0001	91.80 ± 0.28
431/432	IFA-3	1.283 ± 0.010	1.0642 ± 0.0040	95.70 ± 0.12
513	IFA-4	1.266 ± 0.009	1.0680 ± 0.0010	95.47 ± 0.21
527	IFA-5	1.264 ± 0.010	1.0690 ± 0.0004	95.89 ± 0.17

For IFA-527 and –513, the thermal experiments were carried out in a controlled atmosphere with argon and helium in equal proportions. During the test, the heating rate was 150 K/h up to 723 K, then increased to 300 K/h until reaching 1973 K. All samples were held at 1973 K for 24 h and then cooled at a rate of 400 K/h.

All densities were determined by the water immersion technique.

Table 4 - Results of simulation and comparison with experimental data from IFA experiments (Cunningham et al., 1981; Hann et al., 1977)

Fuel Type	Thermal Treatment	$\Delta\rho/\rho_0$		$-3\Delta d/d_0$	
		Exp.	Sim.	Exp.	Sim.
IFA-1	1873 K – 24 h	2.53 ± 0.40	2.22	2.32 ± 0.46	2.18
	1973 K – 4 h	2.58 ± 0.25	2.67	2.44 ± 0.25	2.62
	1973 K – 7.8 h	3.03 ± 0.34	2.89	2.82 ± 0.29	2.84
	1973 K – 24 h	3.37 ± 0.37	3.35	3.07 ± 0.34	3.28
	1973 K – 48 h	3.75 ± 0.81	3.68	3.46 ± 0.64	3.59
IFA-2	1873 K – 24 h	0.09 ± 0.10	0.17	0.11 ± 0.09	0.17
	1973 K – 4 h	-0.07 ± 0.32	0.10	0.11 ± 0.17	0.10
	1973 K – 7.8 h	-0.18 ± 0.84	0.18	-0.02 ± 0.71	0.18
	1973 K – 24 h	0.02 ± 0.46	0.42	-0.11 ± 0.27	0.42
	1973 K – 48 h	0.05 ± 1.20	0.65	-0.14 ± 0.86	0.64
IFA-3	1873 K – 24 h	0.04 ± 0.02	0.03	0.02 ± 0.04	0.03
	1973 K – 4 h	0.09 ± 0.04	0.01	0.09 ± 0.04	0.01
	1973 K – 7.8 h	0.24 ± 0.04	0.03	0.16 ± 0.04	0.03
	1973 K – 24 h	0.25 ± 0.05	0.09	0.21 ± 0.08	0.09
	1973 K – 48 h	0.39 ± 0.02	0.15	0.32 ± 0.07	0.15
IFA-4	1973 K – 24 h	0.35 ± 0.06	0.53	-	0.53
IFA-5	1973 K – 24 h	0.28 ± 0.34	0.35	-	0.35

A summary of the relative change in density and diameter obtained with DIONISIO FEM for re-sintered samples of the IFA experiments is presented in Table 4. For each heat treatment, the experimental data reported here correspond to the mean value and the standard deviation of the measurements taken on four different pellets with similar microstructures. For treatments at

1973 K during 48 hours, only two samples were used. It can be appreciated that the pellets labeled IFA-1 achieved higher densification than those of the other types, essentially due to the presence of a large number of small pores in their microstructures. In most cases, there is a good agreement between the measured and calculated data.

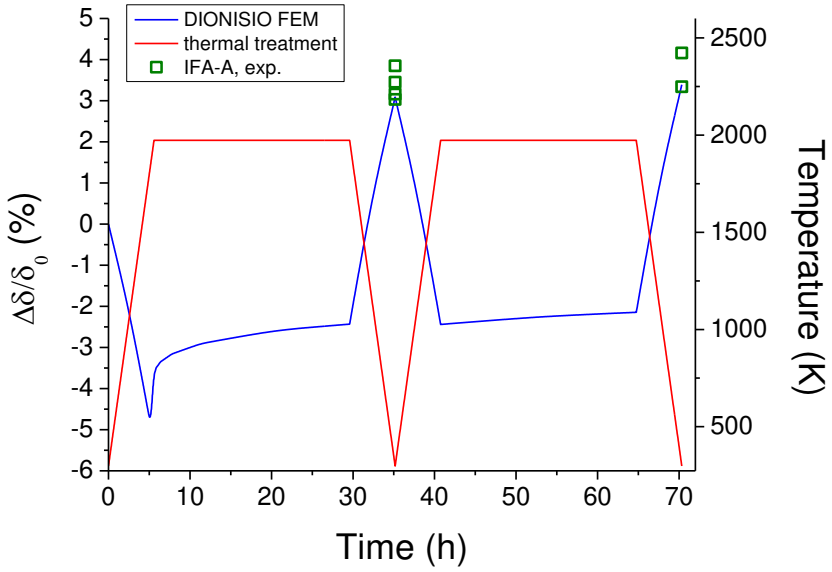

Figure 2 - Density change evolution of the fuel microstructure IFA-1 re-sintered at 1973 K up to 48 h. of isothermal treatment and its comparison with measured valued (Hann et al., 1977).

Figure 2 displays the relative density evolution of an IFA-1 sample during a heating treatment at 1973 K. The thermal history is indicated by the dashed line and consists of two cycles of 24 hours at 1973 K (Hann et al., 1977). During the first heating ramp, the fuel density decreases due to thermal expansion but above 1700 K there is a rapid density increase as a consequence of the accelerated elimination or reduction of pores with a diameter smaller than 1 µm. In the course of the permanence period at 1973 K, the rate at which the material densifies is reduced since larger pores tend to densify less or may even increase in size. Finally, during the cooling ramp, the fuel density increases due to thermal contraction. In the second thermal cycle, the fuel behavior is similar, except that the densification takes place during the stationary stage at a low rate. It is important to notice that the major density increment occurs during the first thermal cycle practically reaching the saturation of the phenomenon. It can be observed that the results obtained with the simulation are within the dispersion range of measured data.

4 CONCLUSIONS

A new module named DIONISIO-FEM was developed in order to simulate out-of-pile experiments and several comparisons were carried out with tests that involved non-irradiated samples under controlled thermal conditions. Particularly, this module was utilized to simulate re-sintering tests of UO_2 pellets with different microstructures and temperature conditions. In most cases, the code has shown a satisfactory agreement with the experimental data, allowing a more accurate and detailed description of the microstructure evolution by incorporating the heating and cooling ramps of a real process.

The new module, supported by the kernel of DIONISIO and its materials library, in continuous progress, extends the capabilities of the code and provides a path to continue improving the simulation of several materials in diverse scenarios of relevance for the nuclear

field.

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