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MODELLING OF FISSION GAS RELEASE IN UO_2 DOPED FUEL USING TRANSURANUS CODE

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Abstract. The expected benefits from Cr-doped fuel are improved retention of fission gases within the pellets due to its large grain size. To demonstrate this, several experiments have been carried out by Halden reactor and Studsvik. These experiments are now being used to benchmark several fuel performance codes among them transuranus code. All this as part of a Coordinate Research Project (CRP) by IAEA named Testing and Simulation for Advanced Technology and Accident Tolerant Fuels (ATF-TS). This work is introducing a novel fission gas diffusivity model for doped fuel in transuranus code. It is observed the benefits of introducing this new model when comparing to the standard model already existing in transuranus. Nevertheless, more work needs to be carried out to fully understand all the phenomena involved in adding dopant in UO_2 due to change of thermo mechanical properties.

Keywords: Doped fuel, transuranus, ATF, fission gas release, diffusivity.

1. Introduction

The study of fission gas release, FGR, is an important area within the fuel rod performance analysis that needs to be covered because these gaseous fission products in the UO_2 fuel pellet are detrimental to the thermo-mechanical properties of the fuel [1]. Fission gas release depends on several mechanisms that occur within the fuel pellet, more specifically within the grain (see Figure 1). Few of these mechanisms are, for instance, thermal contributions: grain boundary diffusion, bubble migration, trapping and resolution; and there are also athermal contributions: recoil, knock-out and sputtering.

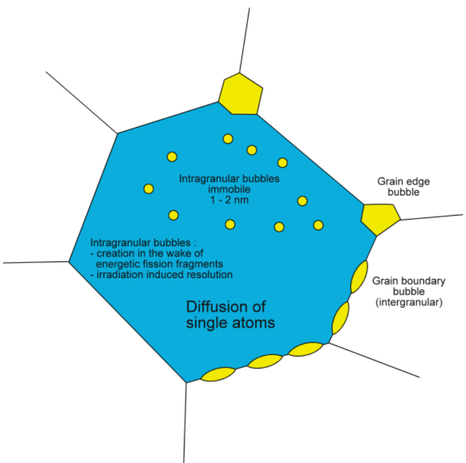


Figure 1. Basic mechanisms of fission gas release [2].

Because in this work we are dealing with doped fuel, it is vital to understand how the properties and characteristics of this fuel will impact on the FGR mechanisms. Although there are other properties that might need to be investigated this work focuses on the selection of a suitable gas diffusivity model for doped fuel.

The selected diffusivity model will be implemented in transuranus code. The new implementation done in the source code will be validated against experimental results performed carried out at the Halden reactor (IFA 716 experiment) and the Studsvik Cladding Integrity Program (SCIP-II). This work is also part of a Coordinate Research Project (CRP) by IAEA[1] named Testing and Simulation for Advanced Technology and Accident Tolerant Fuels (ATF-TS) and the results will be also compared with other members that are simulating this novel fuel in other fuel performance codes.

In Section 2, the methodological details of this work are explained, a brief explanation of the experiments to compare to and considerations taken when performing the simulations. Section 3 shows the results obtained and their comparison against the tests. Finally, in Section 4, it will be shown all the conclusions and also recommendations for future improvements of the modelling of this fuel in transuranus.

2. Methods

The methodology used for the development of this work is as follows i) study the models related to fis-

[1] https://www.iaea.org/projects/crp/t12032

sion gas release in TRANSURANUS (i.e. fission gas diffusivity, thermal conductivity, etc.); ii) select the gas diffusivity model from TRANSURANUS or introduce a new model; iii) verify that the new implemented model works as expected; iv) identify all the fuel properties and reactor conditions required in the input file to run the simulation (type of fuel and cladding, flux, linear power, temperature of the coolant, grain size, etc); v) data collection of the experimental analysis performed by Halden and Studsvik; vi) validate the implemented model by benchmarking the results obtain from TRANSURANUS with the experimental data.

FGR is strictly dependent on temperature; therefore, choosing a good thermal conductivity model for this type of fuel is essential. There is no thermal conductivity model available for doped fuel; however, according to Arborelius et al. there is only a small change in thermal conductivity of fresh fuel due to doping and this small difference is expected to be negligible after the burnup increase [3].Although it would make sense to use standard undoped UO_2 thermal conductivity during simulations the reality is that in this work the correlation used is the one proposed by Ohira et al. for for high burnup UO_2 pellets [4]. The reasoning for this is explained in Section 2.4. TRANSURANUS uses a mechanistic model for fission gas behaviour and release from doctoral thesis of [5]. For the intragranular diffusion coefficient, TRANSURANUS has different models; nevertheless, the model that is most widely accepted for fission gas diffusivity is from [6] which will be named as a standard during this article.

2.1. Selection and modelling of FG diffusivity

Turnbull model for the single gas atom diffusion coefficient can be seen from Equations 1 to 4.

$$D_{eff} = D_1 + D_2 + D_3 \quad (1)$$

$$D_1 = 7.6 \times 10^{-10} e^{-\frac{3500}{T}} \quad (2)$$

$$D_2 = 3.22 \times 10^{-16} \sqrt{R} e^{-\frac{13800}{T}} \quad (3)$$

$$D_3 = 3.22 \times 10^{-16} R, \quad (4)$$

where D_{eff} is the effective diffusion coefficient, D_1 is the intrinsic high-temperature component, D_2 is the irradiation enhanced thermal component, D_3 is the athermal term and R is the rating in W/gU.

Because Che et al. showed a good agreement with Halden FGR experiments when using an empirically tuned enhanced gas diffusion, Cooper et al. decided to investigate in deep the influence of dopant on fission gas diffusivity. In that work, Cooper et al. proposed a diffusion coefficient for uranium doped fuel [7]. Equation 5 shows enhancement of the diffusivity, using the undoped UO_2 diffusivity model in Equation 1 as a baseline.

$$\begin{aligned} D^{doped} = &\exp\left(\frac{H_1}{k_b}\left[\frac{1}{T} - \frac{1}{T_1}\right]\right) D_1^{undoped} + \\ &\exp\left(\frac{H_2}{k_b}\left[\frac{1}{T} - \frac{1}{T_2}\right]\right) D_2^{undoped} + \\ &D_3^{undoped}, \end{aligned} \quad (5)$$

where k_b is the Boltzmann constant. H_1, H_2, T_1 and T_2 are parameters derived for the multiscale analytical enhanced diffusivity model (see Table 1). $D_1^{undoped}$, $D_2^{undoped}$, and $D_3^{undoped}$ are calculated with Equations 2 to 4. Because there are a number of uncertainties when obtaining these parameters Cooper et al. examined two cases namely case A and case B. The first case has minimal alterations migration energy and sink strength given by the oxygen potential. In contrast, case B overestimates this migration energy and sink strength based on the experience from [8].

Parameter	**Case A**	**Case B**
$T_1 = T_2$ [K]	1773	1773
ΔH_1 [eV]	0.3198	0.3282
ΔH_2 [eV]	-0.3345	-0.6998

Table 1. Parameters for the enhanced diffusivity model derived for Cr-doped fuel [7].

Now, it is important to understand the distinction (if any) between the models before implementing the proposed FG diffusivity for the doped fuel in TRANSURANUS. Hence, by plotting the FG diffusion, it is clearly depicted the difference between them (see Figure 2). For the model of the FG diffusivity of doped fuel, the two cases proposed by [7] are considered and they are shown case A and case B.

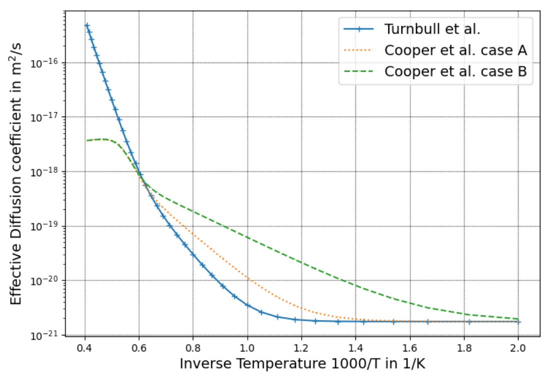


Figure 2. Gas diffusivity in undoped UO_2 fuel using Turnbull et al. model, and gas diffusivity in Cr-doped UO_2 fuel applying Cooper et al. model.

From Figure 2, it is observed that from 700 to 1600 K there is a major difference between case A of Cooper model and the standard diffusion model, whereas for the case B the range is wider from 500 to

1600 K. The ratio between the doped fuel model and the standard model can be as high as 3.4 and 17.5 for case A and B respectively, at around 1000 K. This ratio difference is significant; however, if we consider the range of temperature from 500 to 2000 K the mean ratio will be for case A a factor of 1.63 and for case B a factor 4.66.

2.2. Implementation and verification

In this work both cases of Cooper model are considered. Therefore, it was necessary to have two new versions of TRANSURANUS that includes case A and B in which the only difference is the parameters in Table 1. The implementation done in TRANSURANUS was revised by TRANSURANUS developers to ensure that the coding is up to their standards and minor modifications of the code were proposed by them in terms of clarity of the code.

As evidenced in Figure 2, the value of diffusion coefficient of doped fuel is significantly larger over the range of from 500 to 1600 K, this would mean that if the size of the grain remains the same one would expect a greater FGR when comparing to standard UO_2 fuel. Hence, for the verification of the modelling, several tests were performed. In these tests the grain size was kept the same but using the new model implemented in TU. Table 2 shows the percentage of fission gas release inside the fuel rod and the internal rod pressure difference when using Turnbull model and Cooper model in an another Halden case (IFA 429).

Test cases	Parameter	Model	Value
IFA420cd	FGR	Turnbull	12.34 %
		Cooper	23.17 %
	Pressure	Turnbull	6.45 MPa
		Cooper	7.27 MPa
IFA420ch	FGR	Turnbull	17.79 %
		Cooper	42.95 %
	Pressure	Turnbull	6.95 MPa
		Cooper	11.18 MPa

Table 2. Model verification using IFA 429 case.

For both cases, there is a considerable increase in FGR and pressure when using the diffusivity model for doped fuel. This could confirm that the implementation was done correctly and that the benchmark of the code with experimental data can be carried out.

2.3. Experimental data

The implemented model in TRANSURANUS is validated with experimental data performed by Halden and Studsvik. From the experimental data and fuel characteristics all the input data required is gathered to run the simulation. Some of the input data are unchanged overtime while other data are constantly changing as it is the case of the the power, fast flux and temperature of coolant. For this variable data, an additional processing is needed because TRANSURANUS have a maximum number of record for the Macro-step that can be included in the input file. This limit is up to 5000, however, the data steps provided is around 160000 for IFA 716 and 24000 for SCIP-II. Hence, a software called Fuel Rod Analysis ToolBox (FRAtoolbox) is used to merge the quantities of the large number of data sets [9].

2.3.1. Halden experiment IFA 716

The main objective of the IFA 716 experiment is to investigate the effect of dopant concentration and grain size on the FGR [10]. The experiment was carried out between January 2011 and May 2015 for a total operation time of 842 days divided in 15 operation cycles. IFA 716 assembly consisted in six rods each with different type of fuel.

Few of these characteristics for the IFA 716 case (rod 1 and 2) are reported in Table 3; clearly, the main differences between these two fuels are the Cr_2O_3 content and the fuel average grain diameter. Moreover, there are some pellets that have a hollow section this only applies for the pellets on the top where instrumentation was inserted to obtain, for instance, the on-line measurement of the fuel temperature center line. Figure 3 shows the average linear heat rate measured for the rods 1 and 2 versus time [10]. Evidently, rod 1 has a total operation time of 840 days while rod 2 only has over 700 days. This difference is due to a malfunction in rod 2 that caused this rod to be removed from the reactor, in contrast, rod 1 (and the others) remained until the end of the test. The average linear heat rate (LHR) of both rods are roughly the same throughout the experiment; therefore, the average LHR of rod 1 will be used during the simulation in TRANSURANUS.

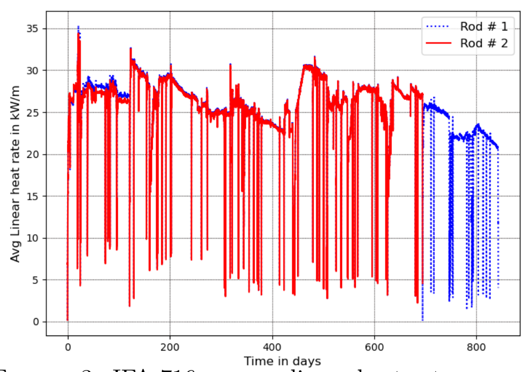


Figure 3. IFA 716 average linear heat rate measured during the test.

2.3.2. Studsvik experiment SCIP-II

The main objective of SCIP-II experiment is to investigate the effect of power ramps on doped fuel with

	IFA 716 Rod 1	IFA 716 Rod 2
Cladding material	Zircaloy-4	Zircaloy-4
Fuel material	$UO_2 + Cr_2O_3$	UO_2
Fill gas	He	He
Pellet outer diameter [mm]	9.12	9.13
Pellet inner diameter, hollow section [mm]	1.8	1.8
Cladding outer diameter [mm]	10.75	10.75
Cladding thickness [mm]	0.725	0.725
Active fuel length [mm]	399.5	399.4
Free volume [m^3]	5.8	5.95
Fuel density [g/cm^3]	10.50	10.55
Fill gas pressure [MPa]	1	1
Fuel Cr_2O_3 content [ppm]	1580	N/A
Fuel U-235 enrichment [wt %]	4.9	4.89
Fuel average grain diameter [µm]	71	11

TABLE 3. Fuel characteristics of IFA 716 rod 1 and 2 simulated in this work [11].

a based irradiation. This based irradiation was carried out between September 2006 and August 2009 at the OKG2 reactor divided in 3 cycles. Similarly, as the IFA 716 case, the fuel characteristics for the SCIP-II case are reported in Table 4. Rods L707 and L708 are fundamentally the same, there is no major distinction between the two according to the given data. What makes these rods different is the base irradiation that they underwent before being subjected to power ramps in a test reactor. Both rods have a total operation time of 1095 days as based irradiation with an average LHR of 14 and 13 kW/m for rod L707 and L708, respectively. This led to a mean BU of 28 MWd/kgU in the case of L707 and 26 MWd/kgU for L708.

The ramp test for each rod was carried out differently for both, with rod L707 having a total time of 21.33 h while rod L708 with a total time of 14.32 h. The ramp was hold for 12 h and around 1 minute for rod L707 and L708, respectively. Figure 4 depicts the average LHR during the ramp test for each rod. In contrast to the Halden experiment, there is no in-situ measurement of fuel center line temperature or plenum pressure which will make the implemented model harder to validate against this experiment. The only measurement that is given comes from the post-irradiation examination where the data for FGR is obtained.

2.4. Additional considerations

The only criteria for selection thermal conductivity model is how close the fuel center line temperature (FCLT) obtained from TRANSURANUS is to the experimental data. TRANSURANUS has a set of thermal conductivity models for different fuels including a correlation for the standard UO_2. When using the LWR standard correlation (model already implemented in TRANSURANUS) proposed by [12], it results in the Figure 8 (top) where evidently there is not a good agreement between this model and the experimental data. Therefore, exploring other thermal conductivity models is necessary to continue to the validation of the FGR. The model that best fit to the experimental results is surprisingly a correlation used for high burnup UO_2. Consequently, the correlation for high burnup UO_2 pellets found in TRANSURANUS is the thermal conductivity model of choice for the study of FGR in this work.

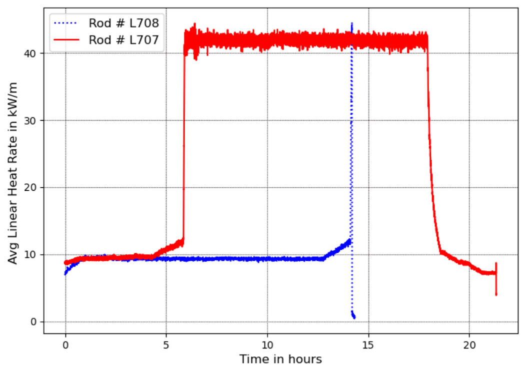


FIGURE 4. SCIP II ramp test.

In addition to using a thermal conductivity for high burnup UO_2, it is considered that the gas diffusivity model for doped fuel is the same independently of the chromia concentration in the fuel because this diffusivity model does not have a correlation as function of the amount of chromia content.

3. Results

The parameters to benchmark are fuel center line temperature, upper plenum pressure, and fission gas release. However, no all data is available for all the rods tested. For instance, for SCIP-II, the only data to compare is the FGR obtained in the post-irradiation examination.

	SCIP-II Rod L707	SCIP-II Rod L708
Cladding material	Zircaloy-4	Zircaloy-4
Fuel material	$UO_2 + Cr_2O_3$	$UO_2 + Cr_2O_3$
Fill gas	He	He
Pellet outer diameter [mm]	8.67	8.67
Cladding outer diameter [mm]	10.05	10.05
Cladding thickness [mm]	0.605	0.605
Active fuel length [mm]	480	480
Fuel density [g/cm^3]	10.50	10.55
Fill gas pressure [MPa]	1	1
Fuel U-235 enrichment [wt %]	3.0	3.0
Fuel average grain diameter [µm]	49	49

Table 4. Fuel characteristics of SCIP-II rod L707 and L708 simulated in this thesis.

3.1. Benchmark of Halden IFA 716

Because pressure transducer of rod 2 was damaged, there is no benchmark of this for rod 2. On the other hand for rod 1, the pressure of case A and B for rod 1 is the same during the first 300–350 days of operation and for both cases TRANSURANUS predicted well the pressure when compared to the experimental data. However, after 350 days of operation TRANSURANUS underestimates the pressure when using diffusivity model case A while when using case B the pressure is overestimated (see Figure 5).

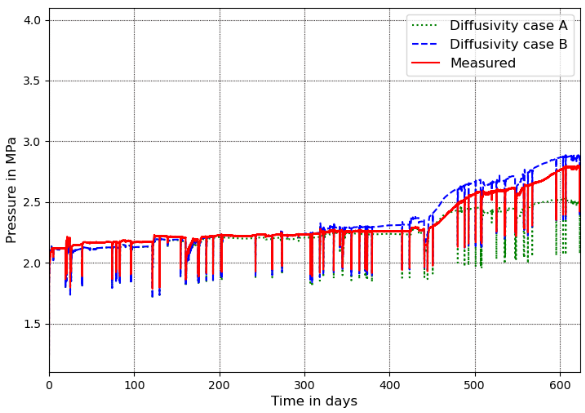


Figure 5. Predicted vs. measured rod pressure for rod 1, including the two cases for the diffusivity of doped fuel.

Figure 7 shows the result of the comparison of the predicted FCLT versus the measured FCLT taken from the test for rods 1 and 2. It can be observed that for Rod 1 (see Figures 7a and 7b), there is a good agreement between the experimental and simulation when the burnup is low enough (< 17 MWd/KgUO_2) while at higher burnup there points that fall out of the established uncertainty zone of ±9 % and yet close to this zone. Around 100 % of agreement is found if we accept this uncertainty. In case of Rod 2, in Figure 7c at low burnup the predicted temperature fall within the uncertainty opposite to what occurs at higher burnup with several data points falling far from this zone. This over-prediction of the temperature, after approximately 400 days of operation, is also observed in Cooper et al. work; however, the temperature difference between predicted and measured is larger [7].

Finally, for the fission gas release, the given data did not contain the evolution of fission gas release versus operation time; instead, what is available is the fission gas release at different stages of the experiment. Nevertheless, this is not true for rod 2, where no data was collected at any point and the rod presented signs of leakage at around 700 days of operation. Hence, no puncture was performed to obtain the fission gas release. Figure 6 shows the predicted fission gas release for rod 1 for both diffusivity cases, the predicted fission gas release for rod 2 and the measures taken by Halden at four different times with their uncertainty for rod 1. Despite having a good agreement between measured and predicted for FCLT and pressure, the fission gas release shows a wide difference between the measured and simulation. This is more evident for rod 1 case A where the prediction of FGR is 1.6 % while case B gives a better results with 3.8 % at 625 days. The measured value for the fission gas release at the same time was 5.71 % ± 1.36 %. Surprisingly, the fission gas release of rod 2 is lower than the rod 1 when using diffusivity case B.

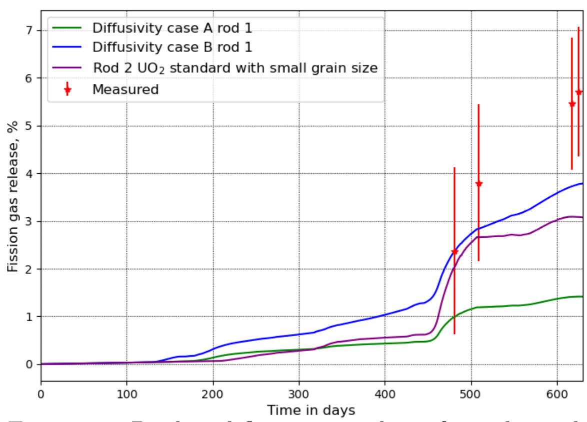
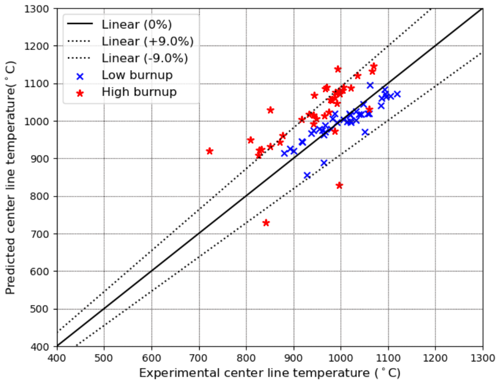
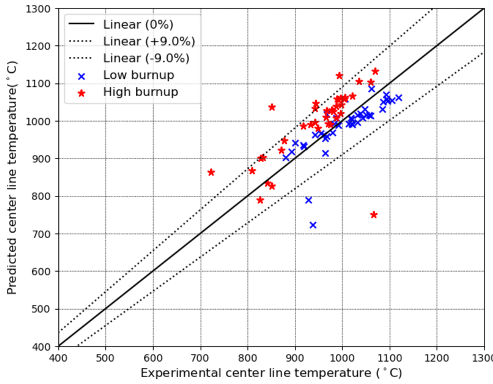


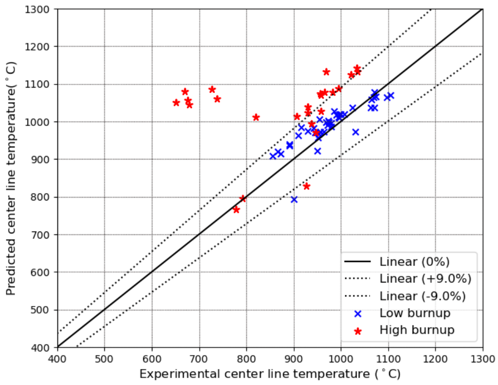
Figure 6. Predicted fission gas release for rod 1 and 2.



(a) . Rod 1 UO_2 doped fuel diffusivity case A



(b) . Rod 1 UO_2 doped fuel diffusivity case B



(c) . Rod 2 standard UO_2 fuel small grain

Figure 7. Predicted vs. measured fuel center line temperature for IFA-716 rods 1 and 2. For burnup lower than 17 MWd/KgUO_2 (shown as '×') and burnup higher than 17 MWd/KgUO_2 (shown as '⋆').

3.2. Benchmark of Studsvik SCIP-II

Unfortunately, there is no data to compare FCLT or pressure as it was done with IFA-716. Table 5 shows the fission gas release obtained during the post irradiation examination and the fission gas release predicted by transuranus using fission gas diffusivity case B. For this experiment, the prediction overestimate the experimental data. This is more evident for the 1 minute ramp (rod L708) where there is almost no release according to PIE but the prediction is 2.5 %.

	Rod L707	**Rod L708**
FGR / PIE	3.0 %	0.04 %
FGR / transuranus	5.8 %	2.5 %

Table 5. Fission gas release comparison at the end of ramp

4. Conclusions

The fission gas diffusivity model was compared to two different experiments, one for ramp test and the other for long period operation. The results show that there are still uncertainties in the prediction using this model in transuranus. The Halden experiment, however, was surrounded by some failures in sensors during the experiment. As a result, it is unclear whether these experimental results can be trusted. Similarly, there was a lack of data to make a better comparison against the SCIP-II experiment. It will be important to continue the validation, perhaps against other experiments, to draw a final conclusion on this model with the transuranus code. Additionally, defining the model exclusively for doped fuel can be beneficial to improving the accuracy of the predictions.

Acknowledgements

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References

[1] M. Tonks, D. Andersson, R. Devanathan, et al. Unit mechanisms of fission gas release: Current understanding and future needs. *Journal of Nuclear Materials* **504**:300–317, 2018. https://doi.org/10.1016/j.jnucmat.2018.03.016.

[2] European Commission. *Transuranus Handbook*, 2019.

[3] J. Arborelius, K. Backman, L. Hallstadius, et al. Advanced doped UO2 pellets in LWR applications. *Journal of Nuclear Science and Technology* **43**(9):967–976, 2006. https://doi.org/10.1080/18811248.2006.9711184.

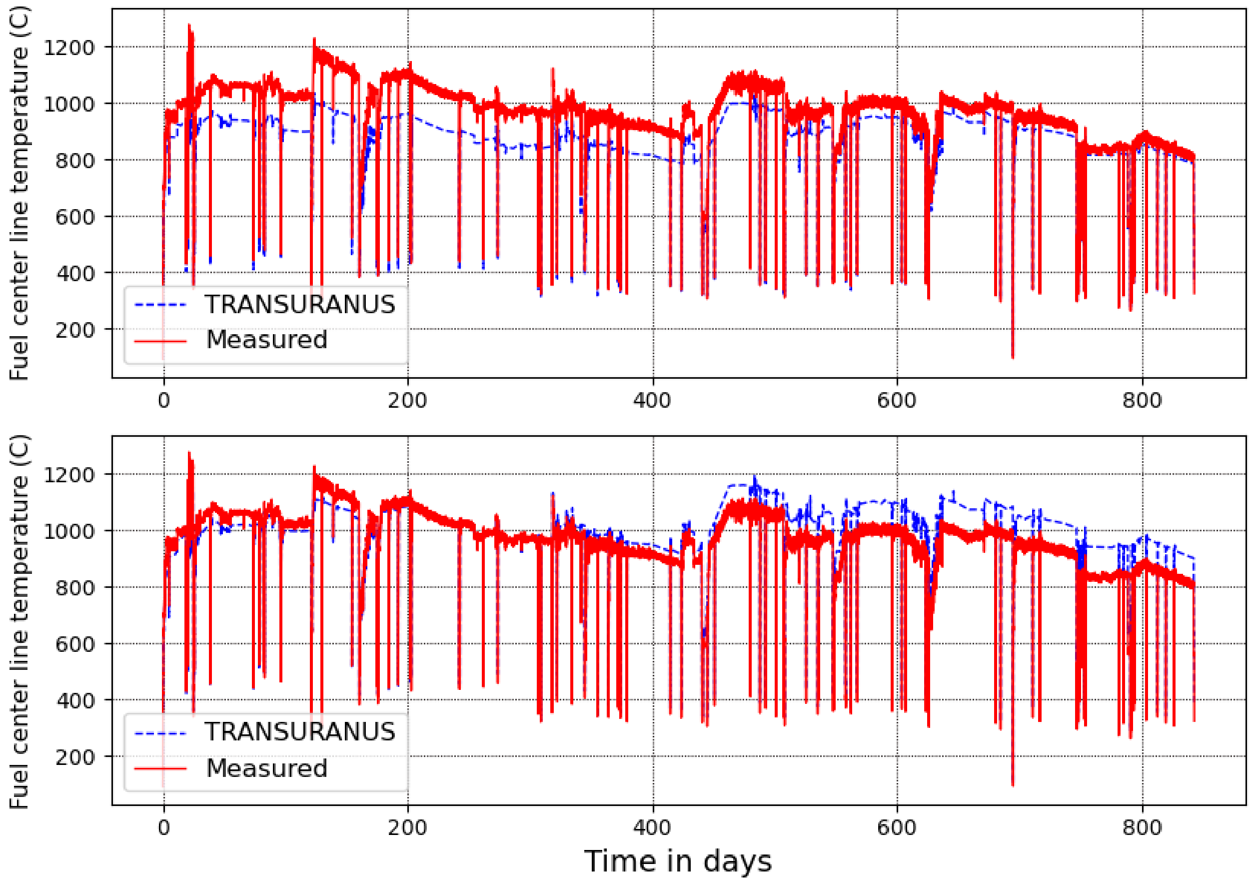

Figure 8. Fuel center line temperature measured by thermocouples during the IFA 716 test compared to simulated FCLT using standard thermal conductivity correlation (top) by Hading et al. [12] and high burnup thermal conductivity correlation by Ohira et al. [4].

[4] K. Ohira, N. Itagaki. Thermal conductivity measurements of high burnup UO2 pellet and a benchmark calculation of fuel center temperature. In *Proceedings 1997 International Topical Meeting on Light Water Reactor Fuel Performance*, pp. 541–549. United States (USA), 1997. ISBN 0-89448-616-0.

[5] G. Pastore. *Modelling of Fission Gas Swelling and Release in Oxide Nuclear Fuel and Application to the TRANSURANUS Code.* Department of Energy, Politecnico di Milano, 2012.

[6] J. Turnbull, C. Friskney, J. Findlay, et al. The diffusion coefficients of gaseous and volatile species during the irradiation of uranium dioxide. *Journal of Nuclear Materials* **107**(2):168–184, 1982. https://doi.org/10.1016/0022-3115(82)90419-6.

[7] M. W. Cooper, G. Pastore, Y. Che, et al. Fission gas diffusion and release for Cr2O3-doped UO2: From the atomic to the engineering scale. *Journal of Nuclear Materials* **545**:152590, 2021. https://doi.org/10.1016/j.jnucmat.2020.152590.

[8] C. Matthews, R. Perriot, M. W. Cooper, et al. Cluster dynamics simulation of uranium self-diffusion during irradiation in UO2. *Journal of Nuclear Materials* **527**:151787, 2019. https://doi.org/10.1016/j.jnucmat.2019.151787.

[9] K. Lassmann, J. van de Laar, A. Schubert, P. V. Uffelen. 'MergeQuantities': a program for adding axial profiles to a {x,y} data set and for merging quantities. Joint Research Centre of the European Commission, 2014.

[10] K. Fuglesang. In-pile results from the fission gas release mechanisms study in IFA-716 after final unloading. Tech. rep., Organisation for Economic Co-operation and Development Halden Reactor Project, Norway, 2016.

[11] M. Sobieska. Production of six fuel rods for fission gas release mechanisms test rig IFA - 716. Tech. rep., Organisation for Economic Co-operation and Development Halden Reactor Project, Norway, 2009.

[12] J. Harding, D. Martin. A recommendation for the thermal conductivity of UO2. *Journal of Nuclear Materials* **166**(3):223–226, 1989. https://doi.org/10.1016/0022-3115(89)90218-3.