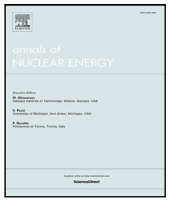
Annals of Nuclear Energy 209 (2024) 110820

Contents lists available at ScienceDirect

Annals of Nuclear Energy

journal homepage: www.elsevier.com/locate/anucene



Consideration of Cr-doped UO_2 fuel performance for a Fluoride-Cooled High Temperature Reactor concept

A. de Lara [a,*], P. Van Uffelen [b], G. Nicodemo [c], G. Zullo [c], D. Pizzocri [c], E. Shwageraus [a]

[a] Department of Engineering, University of Cambridge, Cambridge, United Kingdom
[b] European Commission, Joint Research Centre (JRC), Karlsruhe, Germany
[c] Department of Engineering, Nuclear Reactor Division, Politecnico di Milano, Milano, Italy

ARTICLE INFO

Keywords:
TRANSURANUS
SCIANTIX
Cr-doped fuel
AGR-like FHR

ABSTRACT

This work follows the AGR-like FHR ongoing research that proposes to adopt the British Advanced Gas cooled Reactor (AGR) geometry combined with the molten salt Fluoride-Cooled High Temperature Reactor (FHR) concept.

This work presents the new models and material properties implemented in the TRANSURANUS code for Cr-doped UO_2 fuel, which is one of the advanced technology fuels considered for light water reactors. For this purpose, we update the mechanistic model for fission gas behaviour in the code by means of a dedicated fission gas diffusion coefficient recently proposed by Cooper et al. on the basis of atomistic scale simulations to take into consideration the impact of the dopant on the point defects that control the fission gas diffusivity in various temperature regions of interest. In a consistent manner, we propose also a modified creep correlation based on the mechanistic model for standard oxide fuels. Furthermore, we analyse the effect of cracking observed in doped fuels subjected to power ramps and take into consideration the limited densification of the high-density fuel reported in the open literature. A subsequent parametric study pointed out the main factors affecting the integral fission gas release rate, which was shown to be a limiting factor in the AGR-like FHR under consideration. Finally, the improved performance of the advanced technology fuel is shown by means of the reduced inner gas pressure at end of life, as well as the reduced pellet cladding mechanical interaction during the postulated operational transient from the open literature for AGRs. As a result, the doped fuel is shown to be able to sustain higher power levels in the AGR-like FHR.

1. Introduction

The AGR-like FHR is an innovative reactor concept that proposes to combine the Advanced Gas cooled Reactor (AGR) geometry including its pin type fuel and the molten salt Fluoride-Cooled High Temperature Reactor (FHR) concept (Forsberg et al., 2015).

Previous work on the AGR-like FHR has introduced fuel performance as an integral part of the design space. A new limiting criterion based on the Larson Miller Parameter (LMP) has been proposed. This criterion estimates the time of rupture, maximum cladding temperature and maximum stress that the cladding can sustain using the material dependent LMP equation calculated in a previous work (de Lara et al., 2023). From the LMP, the following ratios are obtained: the ratio of irradiation time over rupture time (cumulative damage function (CDF)); the ratio of maximum stress over maximum cladding stress from the LMP; and the ratio of maximum cladding temperature over maximum cladding temperature from the LMP.

The baseline geometry used in the reference AGR-like FHR is the AGR geometry described in a previous work. The main differences between both reactors are the following: the CO_2 coolant pressure changes from 40 bar to a low pressure molten salt coolant; and Hastelloy-N is used as cladding material instead of stainless steel. The temperature rise across the core is much smaller for the salt, leading to generally higher pin-average coolant temperature for the same core outlet temperature.

Previous work suggests that conventional fuel in the reference AGR-like FHR using the AGR geometry and main parameters could in principle survive an irradiation cycle. However, the fuel pins would experience an increasing internal pressure due to fission gas release, followed by the cladding deforming outwards. This effect is particularly important for the fuel pins in the outermost ring of a fuel assembly which are closer to the graphite moderator (these will be referred to as R3 throughout this paper).

* Corresponding author.
E-mail addresses: ad2048@cam.ac.uk (A. de Lara), Paul.VAN-UFFELEN@ec.europa.eu (P. Van Uffelen), giovanni.nicodemo@mail.polimi.it (G. Nicodemo), giovanni.zullo@mail.polimi.it (G. Zullo), davide.pizzocri@polimi.it (D. Pizzocri), es607@cam.ac.uk (E. Shwageraus).

https://doi.org/10.1016/j.anucene.2024.110820
Received 25 April 2024; Received in revised form 17 June 2024; Accepted 29 July 2024
Available online 7 August 2024

Enlarging fuel grain size has been considered as a promising option to reduce fission gas release. Once the grain boundaries are saturated with fission gas and thermal fission gas release starts, the release rate is controlled by the diffusion of gas from the interior of grains to grain boundaries. Enlarging the size of the grains reduces the influx of fission gas to intergranular bubbles, thereby reducing the release of fission gas through an increase in intragranular diffusion length. Moreover, larger grains can also reduce the intergranular bubble swelling. In order to increase the size of the grains, the temperature for the sintering must be increased, or the oxygen potential must be changed. Such fuels are not commercially attractive. On the other hand, doped fuels as those proposed in some advanced technology fuel (ATF) concepts for LWRs also take advantage of larger grains that can reach initial sizes of 50um (Cheniour et al., 2023).

In line with the current ATF developments, using a Cr-doped UO_2 fuel has been shown to have beneficial effects on the fuel performance. The addition of dopants modifies the microstructure of the fuel during the manufacturing process. The grain size in Cr-doped UO_2 is considerably larger than in undoped fuels.

Fuel performance codes such as BISON (Che et al., 2018) or FRAPCON (GOMES et al., 2021) have already included the models for chromium oxide doped fuel. Material properties for Cr-doped UO_2 have been recently introduced into TRANSURANUS fuel performance code (Lassmann, 1992). In particularly, a new diffusion coefficient, cracking and creep models for Cr-doped UO_2 have been implemented and will be tested in this work. Moreover, the results from the Cr-doped fuel using TRANSURANUS will be compared to the results from the TRANSURANUS-SCIANTIX coupled code with the implemented properties of Cr-doped fuel from Nicodemo et al..

The main objective of this work is to test whether introducing Cr-doped fuel into the AGR-like FHR potentially reduces the amount of fission gas release and improves fuel performance. Moreover, this work will test the newly introduced Cr-doped fuel correlations using the AGR-like FHR reference design as the test case.

2. Methodology

2.1. Configuration

This study is focused on an AGR-like FHR reference model, more specifically, it considers the hottest stringer of a typical AGR. This stringer comprises 36 pins organised into three circular rings, with 6, 12, and 18 pins. The fuel pins are surrounded by graphite sleeves, inner and outer that direct the coolant flow and serve as part of the moderator. Key parameters and design specifications for this assembly are detailed in a previous work (de Lara et al., 2024).

As in a previous study (de Lara et al., 2024), this work focuses on the 5th axial fuel element of the assembly stringer, where the fuel temperature is typically the highest. Particularly on R3, in which the above-mentioned limiting criteria for fuel behaviour phenomena are more extreme because it has highest power due to proximity to the moderator.

Serpent Monte Carlo code (Leppänen et al., 2015) is used to analyse the neutronics. The fuel element and its surrounding moderator geometry were modelled in Serpent (Fig. 1) to calculate the linear heat rate (Fig. 2); and the evolution of the multiplication factor during burnup. The analysis used the JEFF-3.1.2 (Plompen et al., 2020) based cross-section library. Assuming an online refuelling regime, discharge burnup can be approximated as twice that of a single batch core (Kumar, 2016).

2.2. Selection of material properties

Version v1m1j24 (version 1, modification 1 of year 2024) of the TRANSURANUS (TU) code is used to analyse the performance of the fuel. TRANSURANUS is capable of analysing a range of fuel rod conditions using a "1.5D" approach (Lassmann, 1992), Magni et al. (2021). TRANSURANUS also contains material properties and models for Hastelloy-N cladding and FLiBe coolant specific to the AGR-like FHR (de Lara et al., 2023). The analysis assumes a constant outlet temperature within the assembly, and employs a coupling process to integrate neutronics, thermal hydraulics, and fuel performance. The process aims to achieve a more realistic power distribution amongst rings during the initial burnup step.

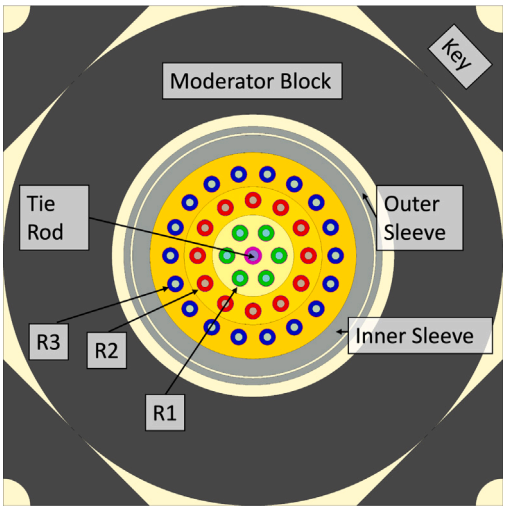


Fig. 1. Cross section of the AGR-like FHR stringer built using Serpent.

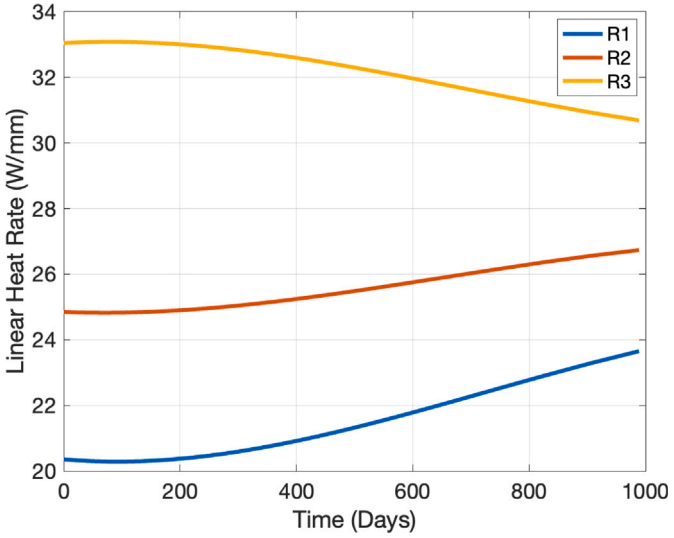


Fig. 2. Linear heat rate in each of the rings of pins in assembly element 5 of the AGR-like FHR.

Moreover, TRANSURANUS allows the coupling with SCIANTIX, which is an independent meso-scale module that provides a more detailed and comprehensive description of the fission products behaviour. This code can be used stand alone, or coupled to other integral thermal-mechanical fuel performance codes. The coupling between TRANSURANUS and SCIANTIX (TU-SC) (Zullo et al., 2023) has already been developed. TRANSURANUS calculates the behaviour of the fuel and cladding, and SCIANTIX calculates fission gas release and fuel swelling (Zullo et al., 2022) using the FISPRO3 model as the driver in the TRANSURANUS code to calculate the fission gas behaviour and swelling. The coupled version improves the conventional, more empirical correlation based methodology used by TRANSURANUS.

For this work, the material properties to model Cr-doped UO_2 fuel were included in TRANSURANUS. These properties are: the diffusion

coefficient based on the work from Cooper (Cooper et al., 2021), a new creep model developed following the work from Malygin et al. for UO_2 (Malygin et al., 2009) and MOX fuel (Malygin et al., 2010a); and Massih and Jernkvist (2015), Cooper et al. (2021) and Dugay et al. (1998) for the Cr-doped fuel. We outline and discuss these properties in the following subsections, along with the specific models and approximations applied for cracking and densification of Cr-doped fuel.

2.2.1. Gas diffusion coefficient

To calculate the effective diffusion coefficient for Cr-doped fuels, a new correlation has been implemented that is based on atomic scale calculations according to Cooper et al. (2021). The diffusion coefficient D^{Doped} is a modified version from that of Turnbull et al. (1982) for undoped fuels where $D^{\text{Undoped}} = D_1 + D_2 + D_3$, represent the intrinsic, irradiation-enhanced, and athermal contributions respectively to fission gas diffusivity. The contribution of D_3 is not affected by the addition of dopants. In accordance with Cooper et al. (2021), D_1 and D_2 are modified to account for the Cr doping of the fuel as follows (Cooper et al., 2021):

$$D^{\text{Doped}} = e^{\left(-\frac{\Delta H_1}{k_B}[1/T-1/T_1]\right)} D_1^{\text{Undoped}} + e^{\left(-\frac{\Delta H_2}{k_B}[1/T-1/T_2]\right)} D_2^{\text{Undoped}} + D_3^{\text{Undoped}} \quad (1)$$

where the parameters T_1, T_2, ΔH_1 and ΔH_2 are reported in Table 3 from Cooper et al. (2021) and k_B is the Boltzmann constant.

Because the gas atoms in the fuel matrix cannot contribute to diffusion, the apparent diffusion is reduced. This difference is considered by an effective diffusion coefficient D_{eff} (Dickon and Nuclear Power training Center, Berkeley Nuclear Laboratories, 2017). The effective diffusion coefficient $D_{\text{eff}} = D^{\text{Doped}} \frac{b}{b+g}$, where g and b are the trapping (Ham, 1958) and resolution (White and Tucker, 1983) parameters respectively:

$$g = 4\pi R N D \quad (2)$$

$$b = 3.03\varphi\pi I_f (R + Z_0) \quad (3)$$

where I_f is the length of a fission fragment track, Z_0 is the radius of influence of the fission fragment, φ represents the flux during the time step under consideration, the bubble radius is R and the bubble density is N.

SCIANTIX uses a different approach to evaluate the diffusion coefficient for the Cr-doped fuel. As for the diffusion coefficient from Cooper et al. the starting point is the diffusion coefficient from Turnbull et al.: $D^{\text{Undoped}} = D_1 + D_2 + D_3$. However, in this case, D^{Undoped} is modified by adding an extra term D_4 to account for the uranium vacancy excess in accordance with point defect modelling (Nicodemo et al., 2024).

Results from Killeen (1980) and Kashibe and Une (1998) indicate that Cr-doped fuel can exhibit higher fission gas diffusivity compared to undoped UO_2. This is the result of higher concentrations of uranium and oxygen vacancies due to more oxidising conditions at high temperatures, and more reducing conditions at low temperatures (Cooper et al., 2021).

Fig. 3 shows the difference between the coefficients from Cooper et al. for doped fuel implemented in TRANSURANUS, and the diffusion coefficient implemented in SCIANTIX (FISPRO3).

2.2.2. Fuel creep

The creep rate is strongly affected by the addition of dopants (Massih and Jernkvist, 2015). The Cr_2O_3 dopant increases the fuel thermal creep rate. A new semi-empirical correlation is developed to model creep in Cr-doped fuels. The correlation relies on a theoretical foundation and aligns with the creep behaviour observed in the UO_2 and MOX models proposed by Malygin et al. (2009, 2010a) (and Liu et al. (2018) for U_3Si_2 fuel). These models agree with the standard creep

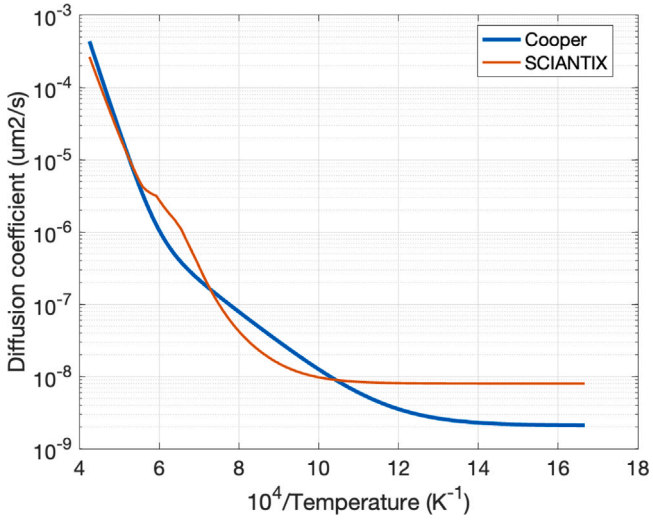


Fig. 3. Modified diffusion coefficient from Cooper et al. for Cr-doped fuel used in TU; and diffusion coefficient from SCIANTIX for Cr-doped fuel.

correlation that TRANSURANUS uses for UO_2 fuel from Lassmann and Moreno (Labmann and Moreno, 1977). A combination of irradiation induced creep and thermal creep terms is considered. More precisely, within the experimental scatter, the steady-state creep rate results from the sum of three terms: a linear function of the stress for low pressure; a power-law dependence with exponent 4.5 at higher pressure; and an athermal term depending linearly on stress.

$$\dot{\varepsilon} = A(\varphi, d)\sigma^{4.5} e^{-Q/RT} + \frac{A_1(\varphi, d)\sigma}{G^2} e^{-Q/RT} + B\sigma\varphi \quad (4)$$

where d is the density of the fuel, σ stands for the effective stress and G represents the grain size.

Malygin et al. (2009) reviewed experimental data and observed that the creep is a linear function of the stress below 30–40 MPa, while a power-law dependence with an exponent of 4–5 is observed for higher pressures. They observed that in the linear range, the creep rate is inversely proportional to the square of the grain size which is also consistent with Eq. (4). Malygin et al. evaluated the activation energy of the thermal creep to be close to that of uranium diffusion by means of a vacancy mechanism. Malygin et al. concluded that, at low stress, creep is controlled by the diffusion mechanism; and at high stress by dislocation climb. They also concluded that both processes operate in parallel and are linearly dependent on the diffusion coefficient, controlled by that of uranium in the ionic compound. They proposed an expression of the diffusion coefficient which takes into consideration the effect of stoichiometry deviations. The result was a thermal creep formulation which depends on temperature, stress, grain size, porosity (P) and deviation from stoichiometry (x). The activation energy on the first term of Eq. (4) was estimated to be 0.5 eV (Malygin et al., 2009), from the energy interaction between a point defect and the dislocation during the climb. The thermal creep for UO_2 is, therefore, as follows:

$$\dot{\varepsilon}_{\text{th}} = (1 + 0.3P^{1.8}) f(x, T) \left[\frac{A\sigma}{G^2} + B\sigma^{4.5} e^{-0.5/k_B T}\right] \quad (5)$$

where A is a constant.

Furthermore, Malygin et al. (2010b) proposed two irradiation induced creep rate components. A pure athermal component controlled by the blocking of interstitial atoms by dislocations; and a thermal component related to the diffusion of point defects towards dislocations loops or clusters, whose orientation depends on the stress state. The former is proportional to the stress, the rate of formation of point defects associated with displacement cascades (K), and inversely proportional to the elastic modulus. The latter is proportional to the stress, the

square root of K and the exponential of the (negative) activation energy Q divided by the temperature in an Arrhenius type equation .

In the same publication (Malygin et al., 2010b), as well as in a specific paper for the thermal creep of MOX fuel (Malygin et al., 2010a), Malygin et al. proposed a semi-mechanistic model for MOX fuel. The irradiation induced components have the same form as for UO_2 fuel (using different parameters); and the components do not depend on the Pu content. The model for thermal creep in MOX fuel on the other hand considers all effects outlined before, in addition to the effect of the Pu concentration and a different consideration of the fuel density.

In order to introduce a Cr-doped fuel creep correlation, it is assumed that the dopants do not affect the irradiation induced term of fission gas, which is also controlled by cation vacancies (according to Cooper et al. (2021)). Cr only affects the thermal creep (as for Pu in MOX fuels). In commercially applied Cr-doped UO_2 fuels, the chromium concentration ([Cr]) is suggested to be between 500 and 2000 wppm (Nelson et al., 2020) (around 0.16 wt%). For this reason, and because of the very limited experimental data available in the open literature, the [Cr] dependency is not considered and is kept constant.

For the thermal creep components, either the temperature dependent correction terms of Cooper et al. applied for Xe diffusion can be considered (Eq 25 in Cooper et al. (2021)); or the model proposed by Massih and Jernkvist (2015). The former proposes a temperature dependent correction term at high temperatures for fission gas diffusion that is controlled by uranium vacancies; and the latter proposes a function of [Cr] for the cation interstitials.

To be consistent with the correlations for UO_2 and MOX from the work of Malygin et al. as well as Cooper et al. the following assumptions are considered:

- Thermal creep in Cr-doped fuel is controlled by the uranium vacancy concentration. Thermal creep is therefore proportional to the corresponding diffusion coefficient that depends on temperature.
- Thermal creep contains a term controlled by U-vacancy diffusion, linearly dependent on the stress and inversely proportional to the grain size; and a term controlled by dislocation climb that contains a power-law dependence for the stress.
- Irradiation creep contains two terms: an athermal component and a thermal component. None of the components of the irradiation creep depend on the Cr content.

The first component of the thermal creep correlation is based on the work of Malygin et al.. This component then relies on the more recent work from Massih and Jernkvist (2015) proposing a modified Nabarro Herring expression for the temperature dependency. Considering that Malygin et al. fitted the coefficients (of an Arrhenius-like function) for thermal creep in UO_2 and MOX fuel based on the experimental data, and also Cooper et al. obtained an Arrhenius-type multiplication factor for the (fission gas) diffusion coefficient, the coefficients for the two thermal creep components of Cr-UO_2 can also be fitted as follows:

$$\dot{\varepsilon}_{\text{Cr-UO}_2\ \text{Thermal}} = \left[\frac{A_3 D_V(T)\sigma}{TG^2} + C D_V(T)\sigma^{5.08}\right] e^{\left(-\frac{\Delta H_1}{k_B}[1/T-1/T_1]\right)} \tag{6}$$

where A_3 and C are fitted to the experimental data of Dugay et al. (1998); the exponent of the power law increases to 5.08 as shown by Dugay et al. (1998) for 0.16wt%. The exponential term is identical to the correction to the diffusion since it is connected to the uranium vacancy diffusion coefficient for the Cr-doped fuel $D_V(T)$. This coefficient is as follows:

$$D_V(T) = D_{V0}(T) e^{\frac{E_D}{k_B T}} \tag{7}$$

where $D_{V0}(T)$ is the pre-exponent for intrinsic U intervacancy diffusion coefficient (m^2/s) in UO_2 and E_D is the migration energy for U vacancy (Massih and Jernkvist, 2015).

For the irradiation term, an Arrhenius-like function from the work of Cooper et al. for D_2 is included in the thermal component to account for the increased diffusivity as follows:

$$\dot{\varepsilon}_{\text{Cr-UO}_2\ \text{Irra}} = \left[\frac{A_1 \sigma K_{V_S}(T,\varphi)}{E(T)} + A_2 e^{\left(-\frac{\Delta H_2}{k_B}[1/T-1/T_2]\right)} \sigma \sqrt{K_{V_S}(T,\varphi)} e^{\left(\frac{-Q}{k_B T}\right)}\right] \tag{8}$$

Where $E(T)$ is the elastic modulus; and A_1 and A_2 are the same as for pure stoichiometric UO_2, assuming that there is no impact of the [Cr] on the diffusivity in the athermal regime. The term $K_{V_S}(T,\varphi)$ is the rate formation of point defects associated with displacement cascades. Malygin et al. (2010a) proposes the stationary solution for the formation rate of mobile radiation defects per unit volume:

$$K_{V_S}(T,\varphi) = \begin{cases} \frac{\varphi}{101} & T < 900\ ^\circ\text{C} \\ \varphi\left(1 + \frac{10^{-18}\varphi}{10^{-20}\varphi + \lambda(T)}\right)^{-1} & T \geq 900\ ^\circ\text{C} \end{cases} \tag{9}$$

where the rate at which defects are freed from trapping centres ($\lambda(T)$) can be expressed as follows:

$$\lambda(T) = 10^{11} e^{\frac{-4.5}{k_B T}} \tag{10}$$

The semi-empirical creep correlation is the combination of the thermal and irradiation creep including the porosity dependence from Malygin et al. as shown in the following Eq. (11).

$$\dot{\varepsilon}_{\text{Cr-UO}_2} = (1 + 0.3P^{1.8})\left(\dot{\varepsilon}_{\text{Cr-UO}_2\ \text{Thermal}} + \dot{\varepsilon}_{\text{Cr-UO}_2\ \text{Irra}}\right) \tag{11}$$

As a first approximation based on the experimental data of Dugay et al. (1998), $A_3 = 3 \times 10^7$ and $B = 584\,496$.

2.2.3. Fuel cracking

The effect of cracks also needs to be considered as it will affect PCMI. To account for the cracks in cylindrical pellets rigorously would require considering the exact location and size of every crack and solving a 3D stress–strain problem in each block. Instead, in most 1.5D codes like TRANSURANUS, one simply modifies the material constants, i.e. ensuring a reduction of the elastic constants (Van Uffelen and Pastore, 2020). It is therefore important to assess the number of cracks in the pellets in order to account for the reduction in effective elastic constants. For this purpose, Bruschi and Barani (2014) proposed a model for the number of cracks as a function of the linear heat rate in conventional UO_2 fuel. According to a report from Areva/Framatome (Holm, 2015), Cr-doped fuel showed more fine radial cracks in the outer part of fuel pellets after a power ramp, which would be beneficial as the resulting PCMI load on the cladding would be more evenly distributed.

In this work, a modified version of the cracking model of Oguma (1983) introducing an upper limit is therefore introduced. This model is used as a first approximation due to the higher number of (radial) cracks in doped fuel during high power operation (ramps). For standard UO_2 fuel, TRANSURANUS contains the Bruschi and Barani (2014) model, which calculates the number of cracks as a function of the linear heat rate by means of a curve that saturates around 10 towards higher power levels. The modified cracking model from Oguma (1983) calculates the cracks using a linear dependency on the linear heat rate. The model covers linear powers between 6 kW m^{-1} and 40 kW m^{-1}; and the model sets the threshold for the formation of the first crack at 6 kW m^{-1}.

Fig. 4 compares the different cracking models against the linear heat rate. The modified cracking model from Oguma shows an increasing number of cracks at much lower linear power, and increases at a higher rate than the value predicted by the model of Bruschi & Barani, which is a reasonable representation of the doped fuel after power ramps.

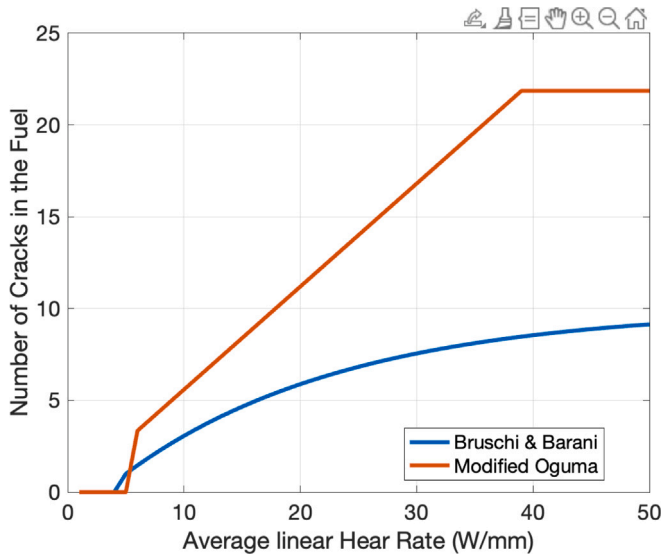


Fig. 4. Number of cracks according to the model of Bruschi & Barani and modified Oguma.

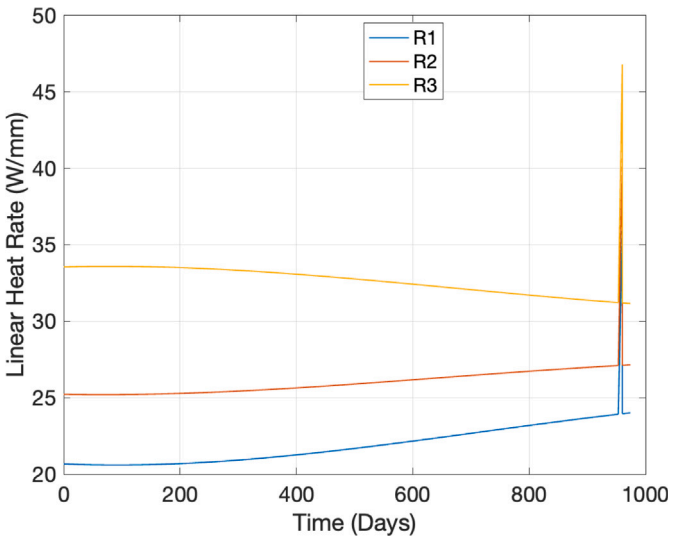


Fig. 5. Linear heat rate in each of the rings of pins in assembly element 5 of the AGR-like FHR showing the power ramp-up.

2.2.4. Fuel densification

According to Massih (2014), introducing additives into the UO_2 fuel leads to higher density fuel, which in turn minimises densification during the irradiation. For this reason, densification is going to be disregarded in the simulations. The lack of densification has an important effect on the fuel outer radius causing the gap size to considerably reduce. The reduced size of the gap affects its conductance lowering the fuel temperature. Therefore, turning densification off has a positive effect as it improves the conductance of the gap, although it will reduce the time for the onset of PCMI.

2.3. Selection of scenario with ramp-up

In order to test the models for the mechanical behaviour of Cr-doped fuel implemented in TU, a ramp-up test is simulated in the reference AGR-like FHR. An example from the work of Haynes et al. (2019) is used as reference, where the linear heat rate increases to 150% for 100 s. The fault leading to the ramp-up takes place at EOL (End Of Life) in Haynes et al. (2019). To test the fuel creep after the increase in power, the ramp-up will be simulated to occur at 99% of the total simulation time. This means that the power increase takes place just before the simulation concludes. Fig. 5 shows the power profile for the ramp-up test.

3. Results

Before showing the results of simulating Cr-doped UO_2 in the AGR FHR-like reactor with the mechanistic version of the fission gas behaviour model in the TRANSURANUS code, the different fission gas behaviour models will be analysed and compared with the previous results. For this analysis, we only consider results for the outer ring R3 as previous studies showed that the fission gas release fraction reaches the highest values in pins in that ring. For the subsequent parametric study in this paper testing the implementation of the Cr-doped correlations, we then made use of the mechanistic version of the fission gas behaviour model of TRANSURANUS. The final results have also been compared against the coupled calculations with SCIANTIX.

3.1. Effect of fission gas behaviour models

The TRANSURANUS code includes different fission product behaviour models: the standard FISPRO model, where swelling is stress dependent, but fission gas release is treated separately; and FISPRO2, a more mechanistic model, where the fission gas release and swelling models are consistent and stress-dependent. Both models use the diffusion coefficient from Matzke (1980), but the mechanistic version of the code uses the effective diffusion coefficient including explicitly the effect of the trapping and resolution rates.

More recently, an alternative to the fission product behaviour models FISPRO and FISPRO2 has been developed in TRANSURANUS. The TRANSURANUS-SCIANTIX coupled code proposes a different fission gas behaviour and swelling models using SCIANTIX, which is called FISPRO3 in this paper.

Fig. 6 shows the differences between models. The fission gas release at EOL using FISPRO2 is considerably lower compared to the basic TRANSURANUS model, despite its shorter incubation period. The TRANSURANUS-SCIANTIX case has the lowest fission gas release fraction, and the incubation period is similar to that of the basic TRANSURANUS. The fastest onset of thermal release in FISPRO2 can be ascribed to the largest intragranular diffusion coefficient at the central fuel temperature around 1200°C(see Fig. 3) during the first 100 irradiation days. Once the grain boundaries reached saturation in FISPRO2, corresponding to the incubation period, the fission gases released to the open gap will deteriorate the thermal heat transfer, which translates into the strong temperature increase and concomitant reduction of the gap conductance as shown in Fig. 7.

Fig. 7 shows that the lowest temperature is predicted by the TU-SC model, which is consistent with the lowest amount of released fission gas. The gap conductance decreases most strongly for FISPRO2 until it starts to grow as the gap closes and the fission gas release reduces. When using the non-mechanistic TRANSURANUS version of the code, the conductance of the gap slightly increases until the onset of fission gas release, when it continues to further decrease. The conductance is predicted to be the highest when using FISPRO3 because of the lowest fraction gas release. Moreover, when using FISPRO2 and FISPRO3, the gap closes, while the gap stays open when using FISPRO, actually revealing a slight tendency to gap reopening at EOL due to the high amount of fission gas release that leads to a large internal gas pressure and a further increasing fuel temperature that could lead to clad rupture due to so-called clad lift-off.

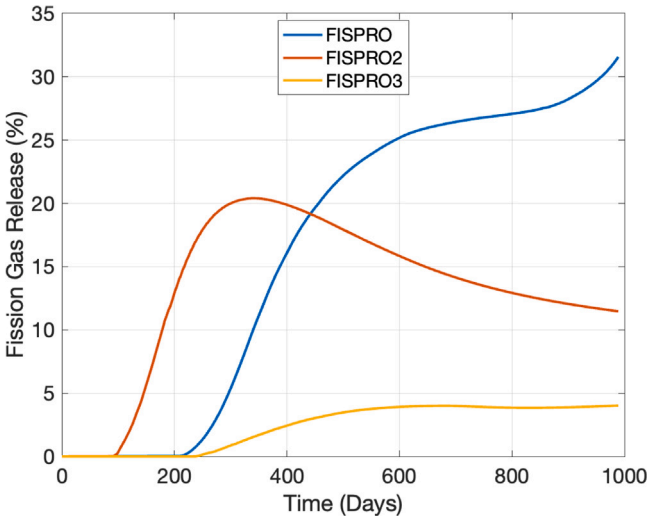


Fig. 6. Fission gas release for the AGR-like FHR using TRANSURANUS FISPRO, TRANSURANUS FISPRO2 and TRANSURANUS-SCIANTIX FISPRO3.

Previous work has used FISPRO to model the behaviour of the fuel as a first approximation since this model was developed for standard fuel. This work will use the mechanistic model (FISPRO2) as new (doped) fuels will be analysed. Moreover, the results will be compared to those obtained with the more mechanistic FISPRO3 model using the TRANSURANUS-SCIANTIX coupled code.

3.2. Parametric study of Cr-doped correlations in TRANSURANUS

A parametric study is performed to assess how the new correlations affect the performance of the (doped) fuel using the AGR-like FHR as the testcase. Among the phenomena in the fuel, fission gas release is the most relevant for this study. Table 1 shows how fission gas release (at EOL) is affected by the Cr-doped specific properties and correlations. The values shown in Table 1 are the fission gas release at EOL for each combination of fuel properties. The Cr-UO_2 diffusion coefficient affects fission gas release the most given the increased mobility of the vacancies in the fuel at the temperatures considered. The effect of the new creep model on the final fission gas release fraction is very minor. The Oguma modified cracking model (Oguma, 1983) causes a modest increase of the fission gas release compared to the previous Bruschi & Barani model (Bruschi and Barani, 2014), while the switching off of the densification has a more important effect on reducing the fission gas release fraction at EOL due to its impact on the gap size, hence the fuel temperatures. From all the specific properties and correlations introduced for doped fuels in TU, the parametric analysis points out that only the diffusion coefficient changes the total fission gas release fraction at EOL beyond the accepted uncertainty band of factor of 2 (Pastore et al., 2015).

3.3. Effect of Cr-doped fuel in the AGR-like FHR during normal operation

Previous work showed the potential improvement that could be achieved in the AGR-like FHR when reducing the fission gas release to the free volume of the pin. For that reason, it was suggested to consider the ATF like for the conventional light water reactors. In order to analyse the resulting improvement, we have implemented the combination of the diffusion coefficient based on Cooper et al. (2021), the newly introduced creep model, the modified cracking model from Oguma (1983) and turned off the densification for simulating the Cr-doped fuel by means of TRANSURANUS and TRANSURANUS-SCIANTIX.

Fig. 8 shows the fission gas released of the undoped fuel and the Cr-doped fuel model using TRANSURANUS and the TRANSURANUS-SCIANTIX coupling. Both calculation schemes show the benefits of Cr doping in terms of reduced fission gas release in the AGR-like FHR as one would expect, and provide results that are within a factor 2 from each other, i.e. they are consistent. Nevertheless, compared to the undoped fuel, the fission gas release is predicted to be lower when using TRANSURANUS. The difference between the undoped and doped cases is less important for the coupled code, which is largely determined by the use of a different diffusion coefficient for the fission gas atoms in the grains as explained above.

Fig. 9 shows the maximum fuel temperature and gap size for the undoped and doped cases. The fuel temperature considerably reduces, most importantly towards BOL where the detrimental effect of fission gas release on the fuel temperatures is most pronounced because of the large gap. The fuel gap grows up to over 30um, while it only reaches 20um for the doped case because of the reduced densification in the doped fuels that are characterised by a larger initial density. This contributes to reducing the maximum fuel temperature and therefore the fission gas release as observed in Fig. 8.

3.4. Effect of cr-doped fuel in the AGR-like FHR during ramp-up test

In order to assess the potential benefits from the advanced technology fuel during ramps in AGR-like FHR in a similar way as in LWRs according to Areva/Framatome (Holm, 2015), we also analysed the power ramp suggested by Haynes et al. (2019). Fig. 10 shows the contact pressure in the cladding for the undoped fuel with a regular and a larger grain size, along with doped fuel. After establishment of the hard PCMI at the onset of the power ramp, the contact pressure is the lowest for the Cr-doped fuel case underlining the beneficial effect of the newly introduced creep model that takes into account the increased mobility of cation vacancies at the temperatures of interest according to Cooper et al. (2021).

The cladding fails for the reference case 30 s after the power ramp-up.

Moreover, by using the Cr-doped fuel, the power ramp-up could be increased by 52%, while it could be increased by 50% for the undoped fuel increasing the grain size up to 50um. The reference case could survive the ramp if the final power is reduced 24%.

4. Summary and conclusions

This work extends previous research on the AGR-like FHR, an innovative reactor combining the features of AGRs and the FHR concept. Previous investigations found that the fission gas released to the free volume of the pin caused increasing pressure and cladding deformation that could lead to clad failure. A potential contribution to reducing the amount of fission gas release is increasing the size of the grains in the fuel. Increasing the grain size can be achieved more effectively by introducing dopants into the UO_2 fuel, which is in line with the current trend to introduce advanced technology fuels in LWRs such as Cr-doped fuel.

This work introduces the very first set of correlations required to model the Cr-doped fuel into TRANSURANUS. Subsequently, we assess the impact of using doped fuel in the AGR-like FHR, with a focus on fission gas release. Moreover, the Cr-doped fuel properties are evaluated against the Cr-doped fuel modelled recently in SCIANTIX by using the coupled TRANSURANUS-SCIANTIX code.

Firstly, the mechanistic version of fission gas behaviour (FISPRO2) in TRANSURANUS is used for this work, i.e. the fission gas release and swelling models are consistent and stress-dependent. In previous research, the conventional FISPRO model for standard UO_2 was applied, where swelling is stress-dependent but fission gas release is treated separately. Since this work is using an advanced technology fuel, it requires a more mechanistic approach in order to better model

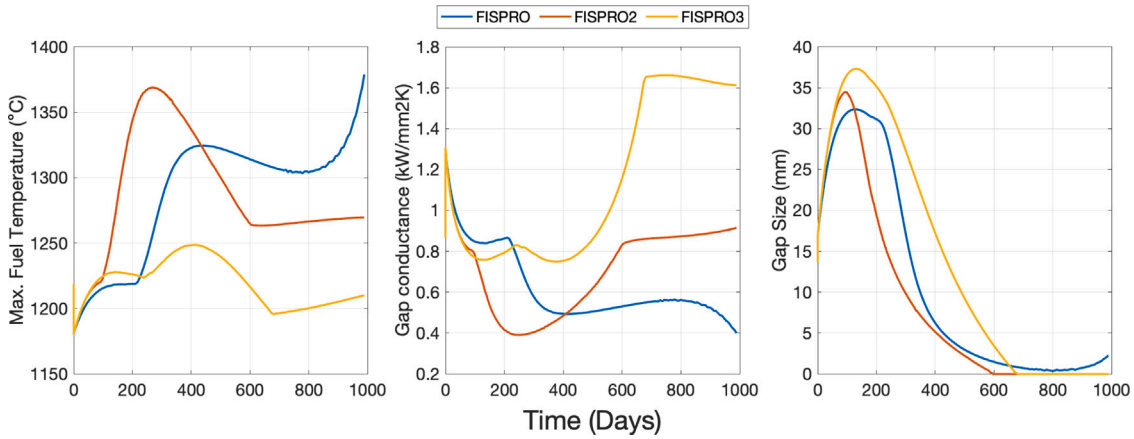


Fig. 7. Maximum fuel temperature, gap conductance and gap size for the AGR-like FHR using TRANSURANUS FISPRO, TRANSURANUS FISPRO2 and TRANSURANUS-SCIANTIX FISPRO3.

Table 1
Fission gas release at EOL (%) for different combination of correlations: densification on/off; cracking off/model from Bruschi & Barani/model from modified Oguma; fission gas diffusion (FG) coefficient from Matzke (UO_2) or from modified Cooper ($Cr\text{-}UO_2$); and creep model for UO_2 from Malygin or the new correlation for $Cr\text{-}UO_2$. The data is obtained from the AGR-like FHR simulations.

		FG Diffusion UO_2		FG Diffusion $Cr\text{-}UO_2$	
		Creep UO_2	Creep $Cr\text{-}UO_2$	Creep UO_2	Creep $Cr\text{-}UO_2$
Densification Off	Crack Off	1.84	1.97	5.71	5.70
	Crack Bruschi & Barani	1.84	1.97	5.71	5.69
	Crack Oguma	1.83	1.95	5.65	5.62
Densification On	Crack Off	2.08	2.63	5.61	6.43
	Crack Bruschi & Barani	2.10	2.66	5.70	6.52
	Crack Oguma	2.12	2.84	6.19	7.37

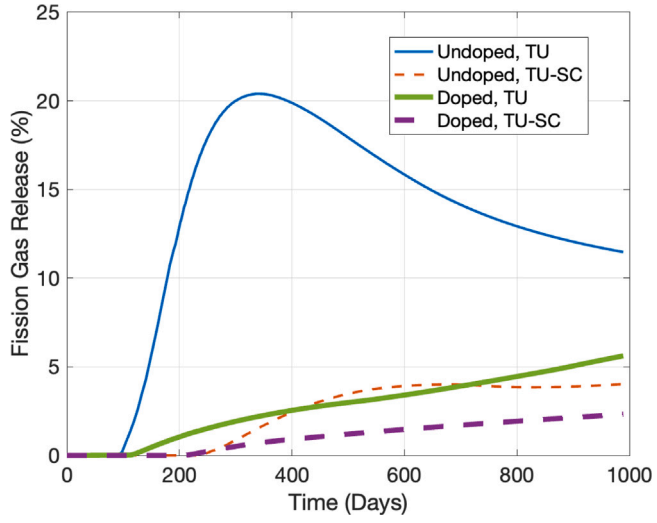


Fig. 8. Fission gas release for the AGR-like FHR using undoped and doped fuel.

its properties. Moreover, SCIANTIX also uses a mechanistic approach by modelling an effective diffusion coefficient. To perform a sensible comparison between both models, the mechanistic version of TRANSURANUS is used in this work.

To model the Cr-doped fuel in TU, at least the following properties are required as pointed out in the literature: the gas diffusion coefficient, the fuel cracking and creep. As a first step in this work, the diffusion coefficient has been developed using a modified version of the coefficient by Cooper et al. starting from the coefficient developed by Turnbull et al.. In addition, the Oguma model has been adapted for cracking in Cr-doped fuels as a preliminary approach, replacing the earlier Bruschi & Barani model for UO_2. According to the Oguma model, the number of cracks is larger at high power compared to the Bruschi & Barani model, in which the quantity of cracks saturates earlier as a function of the linear heat rate. The modified Oguma model provides a more precise depiction of cracking behaviour in Cr-doped fuels as revealed in ramp tests reported by Areva. The creep correlation has been developed for this work based on the semi-empirical developments of Malygin et al. for UO_2 and MOX; the experimental data published by Dugay et al. and the atomic-scale study of Cooper et al. for Cr-doped fuel. Finally, studies have found that there is little to no densification in Cr-doped fuel. For this reason, the densification has been turned off in TRANSURANUS to model doped fuel.

The thermal conductivity of Cr-doped fuel was assumed to be identical to that of undoped fuel. Any differences are minimal and become more noticeable at low temperatures. Therefore, the thermal conductivity for Cr-doped fuel remains unchanged from undoped UO_2 (Arimescu et al., 2019).

A parametric study has been preformed to assess the impact of the introduced first set of correlations for Cr-doped fuel in the AGR-like FHR under consideration. The fission gas release increases most by introducing the modified diffusion coefficient from Cooper et al.

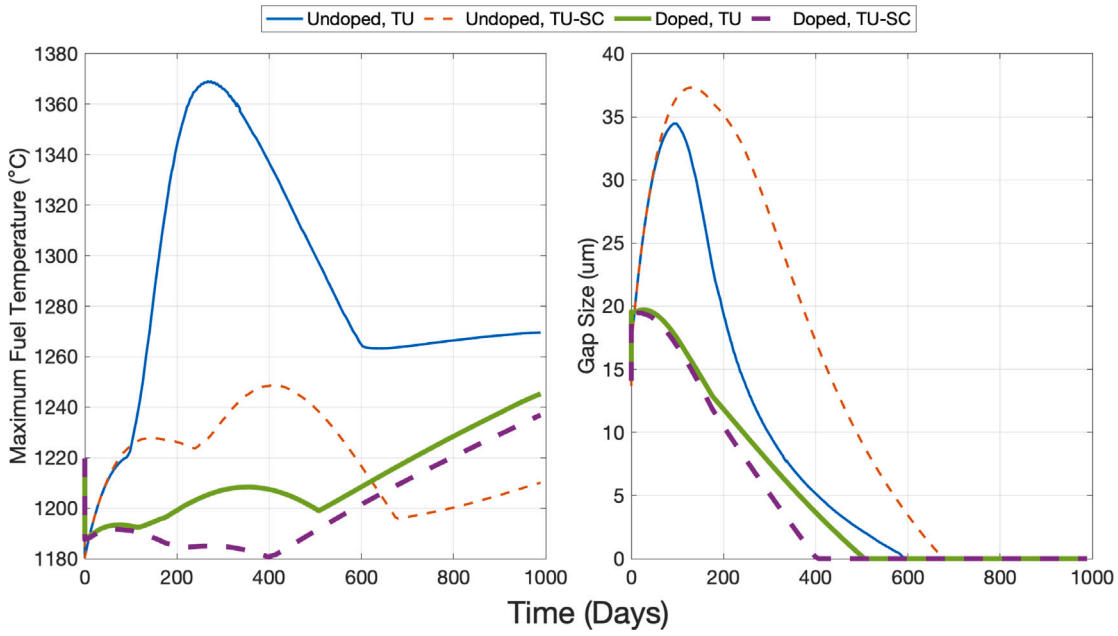
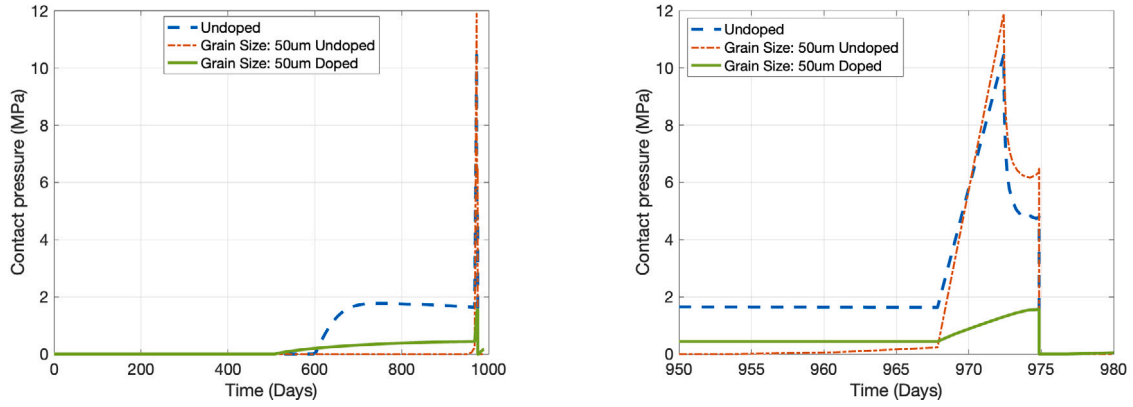


Fig. 9. Maximum fuel temperature, and gap size for the undoped and doped AGR-like FHR.



(a) Contact pressure. (b) Zoom into the contact.

Fig. 10. Contact pressure in the pin using the mechanistic version of TRANSURANUS for the undoped, large grain and doped cases.

since the time required for the fission products to achieve the grain boundaries reduces. The addition of the new creep correlation increases slightly the amount of fission gas as well, due to smaller hydrostatic stress levels in the fuel but it reduces the incubation period mainly because the gap size is affected. Apart from the effect on fission gas mobility due to Cr doping, the most important effect is the lack of densification. The fission gas is largely reduced thanks to a smaller gap size BOL, leading to lower fuel temperatures.

The creep in Cr-doped fuel is larger when compared to the standard UO_2 fuel at the relatively high temperatures experienced in the AGR-FHR, which is directly resulting from the increased thermal creep rate due to Cr-doping of UO_2 fuel according to the new correlation developed in this work. A higher creep rate, in combination with a larger number of cracks in the fuel according to the modified Oguma correlation presented in this manuscript, enables the material to better accommodate applied stress during PCMI, i.e. reduce the clad hoop stress. In order to evaluate this effect, a ramp-up test in an AGR-like FHR taken from the open literature is simulated to evaluate the impact of the new creep correlation. The test showed that the contact pressure is largely reduced for the Cr-doped fuel compared to the undoped fuel with a regular grain size and a large grain size. When accounting for all modified fuel properties, the doped fuel can sustain higher power levels in comparison with standard fuel during the ramp test scenario found in the open literature.

Nevertheless, it is important to highlight that the presented analysis has only been applied to AGR-like FHR conditions using a first version of the TRANSURANUS code for Cr-doped fuel. Definite conclusions would need more experimental validation, especially for irradiated fuel and considering also burnup effects in the cladding under conditions that are representative for an AGR-like FHR as was pointed out in a previous paper.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Paul Van Uffelen reports financial support was provided by Euratom Research and Training Programme. Alejandra de Lara reports financial support was provided by NEF EPSRC. Paul Van Uffelen reports a relationship with European Commission, DG Joint Research Centre that includes: employment. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgements

This work was supported by UK EPSRC Centre for Doctoral Training in Nuclear Energy Futures, grant number EP/S023844/1.

P. Van Uffelen acknowledges funding from the Euratom research and training programme 2021–2027 through the APIS project (https://apis-project.eu/) under grant agreement n° 101114673. Views and opinions expressed in this paper reflect only the authors' view and the Commission is not responsible for any use that may be made of the information it contains.

Co-funded by the European Union

References

Arimescu, V., Davis, T., Korolev, V., Anderson, M., Delafoy, C., 2019. Implementation of chromia-doped fuel in rodex4 and downstream codes and methods for U.S. BWR applications. Top Fuel.

Bruschi, E., Barani, T., 2014. Extension and validation of a transient fission gas release model for the transuranus fuel performance code.

Che, Y., Pastore, G., Hales, J., Shirvan, K., 2018. Modeling of Cr2O3-doped UO2 as a near-term accident tolerant fuel for LWRs using the BISON code. Nucl. Eng. Des. 337, 271–278.

Cheniour, A., Sweet, R.T., Nelson, A.T., Wilson, B.A., Shields, A.E., 2023. Sensitivity of UO2 fuel performance to microstructural evolutions driven by dilute additives. Nucl. Eng. Des. 410, 112383.

Cooper, M.W., Pastore, G., Che, Y., Matthews, C., Forslund, A., Stanek, C.R., et al., 2021. Fission gas diffusion and release for Cr2O3-doped UO2: From the atomic to the engineering scale. J. Nucl. Mater. 545, 152590.

de Lara, A., Pizzocri, D., Van Uffelen, P., Shwageraus, E., 2024. The AGR-like FHR reactor: Assessing the technical limits of the fuel. Nucl. Eng. Des. 423, 113180.

de Lara, A., Van Uffelen, P., Schubert, A., 2023. Towards preliminary design calculations for application of hastelloy cladding material. Ann. Nucl. Energy.

Dickon, I.K., Nuclear Power training Center, Berkeley Nuclear Laboratories, 2017. Transuranus Handbook. Pergamon.

Dugay, C., Mocellin, A., Dehaudt, P., Sladkoff, M., 1998. High temperature mechanical tests performed on doped fuels.

Forsberg, C., Richard, J., Pounders, J., Kochendarfer, R., Stein, K., Shwageraus, E., et al., 2015. Development of a fluoride-salt-cooled high-temperature reactor (FHR) using advanced gas-cooled reactor (AGR) technology. Trans. Am. Nucl. Soc. 112, 566.

GOMES, D.d.S., et al., 2021. Analysis of chromium oxide doped pellet for LWRs using FRAPCON code.

Ham, F.S., 1958. Theory of diffusion-limited precipitation. J. Phys. Chem. Solids 6 (4), 335–351.

Haynes, T., Podgurschi, V., Wenman, M., 2019. The impact of azimuthally asymmetric carbon deposition upon pellet-clad mechanical interaction in advanced gas reactor fuel. J. Nucl. Mater. 513, 62–70.

Holm, J., 2015. Cr 20 3 Doped U0 2 Fuel, BWR Implementation. AREVA/NRC Rockville, MD.

Kashibe, S., Une, K., 1998. Effect of additives (Cr2O3, Al2O3, SiO2, MgO) on diffusional release of 133Xe from UO2 fuels. J. Nucl. Mater. 254 (2–3), 234–242.

Killeen, J., 1980. Fission gas release and swelling in UO2 doped with Cr2O3. J. Nucl. Mater. 88 (2–3), 177–184.

Kumar, C., 2016. Nuclear Fuel Management. Stanford University, http://large.stanford.edu/courses/2016/ph241/kumar2/.

Labmann, K., Moreno, A., 1977. The Light-Water-Reactor Version of the URANUS Integral Fuel-Rod Code. Junta de Energia Nuclear (JEN).

Lassmann, K., 1992. Transuranus: A fuel rod analysis code ready for use. J. Nucl. Mater. 188, 295–302.

Leppänen, J., Pusa, M., Viitanen, T., Valtavirta, V., Kaltiaisenaho, T., 2015. The serpent Monte Carlo code: Status, development and applications in 2013. Ann. Nucl. Energy 82, 142–150.

Liu, R., Zhou, W., Cai, J., 2018. Multiphysics modeling of accident tolerant fuel-cladding U3Si2-FeCrAl performance in a light water reactor. Nucl. Eng. Des. 330, 106–116.

Magni, A., Del Nevo, A., Luzzi, L., Rozzia, D., Adorni, M., Schubert, A., et al., 2021. The transuranus fuel performance code. In: Nuclear Power Plant Design and Analysis Codes. Elsevier, pp. 161–205.

Malygin, V., Naboichenko, K., Shapovalov, A., Bibilashvili, Y.K., 2009. Recommendations for calculating the thermal creep properties of uranium dioxide when analyzing fuel-element serviceability. At. Energy 107, 381–386.

Malygin, V., Naboichenko, K., Shapovalov, A., Bibilashvili, Y.K., 2010a. Recommendations for calculating the characteristics of thermal creep of mixed uranium–plutonium oxide fuel when analyzing fuel-element serviceability. At. Energy 108, 15–20.

Malygin, V., Naboichenko, K., Shapovalov, A., Bibilashvili, Y.K., 2010b. Recommendations for calculating the rate of irradiation-induced creep of oxide fuel when analyzing fuel-element serviceability. At. Energy 108, 121–126.

Massih, A., 2014. Effects of Additives on Uranium Dioxide Fuel Behavior. Strålsäkerhetsmyndigheten (SSM).

Massih, A., Jernkvist, L.O., 2015. Effect of additives on self-diffusion and creep of UO2. Comput. Mater. Sci. 110, 152–162.

Matzke, H., 1980. Gas release mechanisms in UO2—A critical review. Radiat. Eff. 53 (3–4), 219–242.

Nelson, A.T., Silva, C.M., Lupercio, A., Moshkelgosha, E., Winters, R., Doyle, C., et al., 2020. Fracture Strength Determination Methods for Ceramic Materials Applied to Uranium Dioxide. Oak Ridge National Lab.(ORNL), Oak Ridge, TN (United States); Lawrence ....

Nicodemo, G., Zullo, G., Cappia, F., Van Uffelen, P., De Lara, A., Luzzi, L., Pizzocri, D., 2024. Chromia-doped uo2 fuel: an engineering model for chromium solubility and fission gas diffusivity. Journal of Nuclear Materials 155301.

Oguma, M., 1983. Cracking and relocation behavior of nuclear fuel pellets during rise to power. Nucl. Eng. Des. 76 (1), 35–45.

Pastore, G., Swiler, L., Hales, J.D., Novascone, S.R., Perez, D.M., Spencer, B.W., et al., 2015. Uncertainty and sensitivity analysis of fission gas behavior in engineering-scale fuel modeling. J. Nucl. Mater. 456, 398–408.

Plompen, A.J., Cabellos, O., De Saint Jean, C., Fleming, M., Algora, A., Angelone, M., et al., 2020. The joint evaluated fission and fusion nuclear data library, JEFF-3.3. Eur. Phys. J. A 56 (7), 1–108.

Turnbull, J., Friskney, C., Findlay, J., Johnson, F., Walter, A., 1982. The diffusion coefficients of gaseous and volatile species during the irradiation of uranium dioxide. J. Nucl. Mater. 107 (2–3), 168–184.

Van Uffelen, P., Pastore, G., 2020. 2.13 - Oxide fuel performance modeling and simulation. In: Konings, R.J., Stoller, R.E. (Eds.), Comprehensive Nuclear Materials (Second Edition), second ed. Elsevier, Oxford, pp. 363–416, Available from: https://www.sciencedirect.com/science/article/pii/B9780128035818116935.

White, R., Tucker, M., 1983. A new fission-gas release model. J. Nucl. Mater. 118 (1), 1–38.

Zullo, G., Pizzocri, D., Luzzi, L., Kremer, F., Dubourg, R., Schubert, A., et al., 2023. Towards simulations of fuel rod behaviour during severe accidents by coupling transuranus with SCIANTIX and MFPR-F. Ann. Nucl. Energy 190, 109891.

Zullo, G., Pizzocri, D., Magni, A., Van Uffelen, P., Schubert, A., Luzzi, L., 2022. Towards grain-scale modelling of the release of radioactive fission gas from oxide fuel. Part II: Coupling SCIANTIX with transuranus. Nucl. Eng. Technol. 54 (12), 4460–4473.