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Technical Note

On the intra-granular behaviour of a cocktail of inert gases in oxide nuclear fuel: Methodological recommendation for accelerated experimental investigation

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ABSTRACT

Besides recent progresses in the physics-based modelling of fission gas and helium behaviour, the scarcity of experimental data concerning their combined behaviour (i.e., cocktail) hinders further model developments. For this reason, in this work, we propose a modelling methodology aimed at providing recommendations for accelerated experimental investigations. By exploring a wide range of annealing temperatures and cocktail compositions with a physics-based modelling approach we identify the most interesting conditions to be targeted by future experiments. To corroborate the recommendations arising from the proposed methodology, we include a sensitivity analysis quantifying the impact of the model parameters on fission gas and helium release, in conditions representative of high and low burnup.

1. Introduction

The description of intra-granular gas behaviour is typically the first step in physics-based models for the prediction of gas release and swelling in nuclear fuel performance codes [1–4]. Although the separate behaviour of helium and fission gas (xenon and krypton) has been studied in detail, both from the modelling [5–12] and experimental point of view [13–19], scarce attention has been paid to their coupled behaviour. Even the work by Hiernaut and co-authors [20,21], which investigated the simultaneous release of several fission products (including helium and xenon) from irradiated MOX fuel samples, lacks the characterization of the samples required for the connection of their experimental results with physics-based models. The present work represents thus a first modelling attempt to address the coupled intra-granular behaviour of helium and fission gas and is aimed at supporting the development of dedicated experimental activities covering the current gap in available data.

Relying on the state-of-the-art modelling capabilities [7,22,23], we propose a model that derives from the synergy of two different studies that separately involve helium and fission gas. The model describes their coexistence in intra-granular bubbles, the processes exchanging helium and fission gas from the bubbles to the solution, and the diffusion towards the grain boundaries. Without lack of generality, the proposed model is applied in annealing conditions, i.e., the conditions of interest for the realization of experiments investigating helium/fission gas release. The method proposed is to use a physics-based modelling to explore the space of experimental conditions (i.e., composition of the cocktail of helium and fission gas, annealing temperature) to identify those conditions highlighting peculiar and interesting behaviours, enabling to extract the parameters of interest.

The proposed methodological approach, even if constructed on physics-based models, requires the a priori application of a non-validated model. Thus, to further support the recommendations provided, we performed a sensitivity analysis with the Pareto method quantifying the impact of the uncertainties in four model parameters on the model predictions. The sensitivity analysis points out that the diffusion coefficients and Henry's constant have the major influence on the model behaviour prioritising the reduction of their uncertainties to improve the model predictive capability.

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Table 1
Model parameters representative of the intra-granular processes in annealing conditions.

Symbol	Description	Formula[a]	u.o.m.	Reference
$\boldsymbol{D_{FG}}$	Fission gas diffusion coefficient	$D_{FG} = D_1 + D_2 + D_3$[b] $D_1 = 7.6\ 10^{-10}\ \exp(-3.04/k_BT)$	$m^2\ s^{-1}$	[32]
$\boldsymbol{D_{He}}$	Helium diffusion coefficient	$2.0\ 10^{-10}\ \exp(-2.12/k_BT)$	$m^2\ s^{-1}$	[31]
$\boldsymbol{g_{FG}}$	Fission gas trapping rate	$4\pi D_{FG}(R_{ig}+R_{FG})N$ $R_{FG} = 0.21\ 10^{-9}$ m	s^{-1}	[9,34], Section 2
$\boldsymbol{g_{He}}$	Helium trapping rate	$4\pi D_{He}(R_{ig}+R_{He})N$ $R_{He} = 4.73\ 10^{-11}$ m	s^{-1}	[9,34], Section 2
$\boldsymbol{g'_{FG}}$	Fission gas trapping rate per bubble	$4\pi D_{FG}(R_{ig}+R_{FG})$	s^{-1}	[9,34], Section 2
$\boldsymbol{g'_{He}}$	Helium trapping rate per bubble	$4\pi D_{He}(R_{ig}+R_{He})$	s^{-1}	[9,34], Section 2
γ	Helium thermal re-solution rate	$4\pi D_{He}R_{ig}k_H\frac{k_BTZ}{V_{ig}}$	s^{-1}	[7], Section 2
$\boldsymbol{k_H}$	Henry's constant	$4.1\ 10^{24}\exp(-0.65/k_BT)$	at $m^{-3}\ Pa^{-1}$	[27]

[a] T (K) is the temperature and k_B (eV K^{-1}) is the Boltzmann constant.
[b] Only the intrinsic thermal diffusivity is considered since the model is to be applied in annealing conditions (fission rate equal to zero).

2. Model description

The model presented in this section describes the coupled intra-granular behaviour of a cocktail of noble gases[1] in UO_2. A reduced version of the model is proposed that is sufficient for the simulation of the gas behaviour in annealing conditions. The processes involved (Table 1) are gas atom diffusion, thermal re-solution and gas atom trapping at intra-granular bubbles. The model is implemented in SCIANTIX [23,24]. In the final form, it consists of six differential equations, three of them are referred to fission gas behaviour and three to helium behaviour. On the one side, the equations proposed for helium derive from the work of Cognini et al. [7]. The latter in fact has a fission-gas-inspired model structure, that brings about the possibility to explicitly account for the interaction between helium and fission gas. On the other side, the fission gas formulation refers to the single-size model of Pizzocri et al. [8]. The number of bubbles N remains constant since no fission (related to the *heterogeneous* nucleation process [25]) occurs in annealing conditions. The following model is applicable to the simulation of fast annealing experiments performed in vacuum conditions [7]. Thus, we write:

$$\begin{cases} \dfrac{\partial c_{FG}}{\partial t} = D_{FG}\nabla^2 c_{FG} - g_{FG}c_{FG} \\ \dfrac{\partial m_{FG}}{\partial t} = g_{FG}c_{FG} \\ \dfrac{\partial n_{FG}}{\partial t} = g'_{FG}c_{FG} \\ \dfrac{\partial c_{He}}{\partial t} = D_{He}\nabla^2 c_{He} - g_{He}c_{He} + \gamma m_{He} \\ \dfrac{\partial m_{He}}{\partial t} = g_{He}c_{He} - \gamma m_{He} \\ \dfrac{\partial n_{He}}{\partial t} = g'_{He}c_{He} - \gamma n_{He} \end{cases} \quad (1)$$

where the main variables are c (at m^{-3}) the gas grain solution concentration, m (at m^{-3}) the gas grain bubbles concentration, n (at $bubble^{-1}$) atoms per intra-granular bubbles. The subscripts identify fission gas (FG) and helium (He). The diffusivities D ($m^2\ s^{-1}$), the trapping rates g (s^{-1}), and the thermal re-solution rate γ (s^{-1}) are detailed in Table 1 and in the following subsections. Lastly, g' ($m^3\ bubble^{-1}\ s^{-1}$) corresponds to g/N, i.e., the specific trapping rate per bubble.

The intra-granular bubble population is described by a single-size model [8] detailed in this section. Bubbles nucleate instantaneously at the average size and are destroyed according to the heterogeneous mechanism always at the average size. In this specific case, it is assumed that a bubble population is formed at the first time-step and then it remains constant during the whole simulation.

The boundary conditions assumed for the single-atoms diffusion problem are $c(a) = 0$ and $|\partial c/\partial r|_0 = 0$, for both helium and fission gas, where a (m) is the grain radius and r (m) is the radial coordinate along the grain radius. The grain is assumed spherical.

One of the main reasons of the peculiarity of helium behaviour in oxide fuels compared to fission gases comes from its higher solubility [16,17]. The solubility demands a model that includes the thermal re-solution process. This work follows the procedure proposed by Cognini et al. [7] with regard to the derivation of the thermal re-solution. It has been verified that the helium solubility in oxide nuclear fuel is linearly proportional to the infusion pressure at a fixed temperature [16,26,27]. According to Dalton's law of partial pressures, the total pressure p_{ig} (Pa) exerted by a mixture of gases is equal to the sum of the partial pressures of each of the constituent gases in this case. By applying Dalton's law and using the Carnahan–Starling equation of state [28] we obtain the partial pressure of helium as:

$$p_{He} = x_{He}\, p_{ig} = \frac{k_BTZ}{V_{ig}} n_{He} \quad (2)$$

where V_{ig} (m^3) is the intra-granular bubble volume and Z (/) is the compressibility factor.

The bubble radius is a key parameter for the prediction of the intra-granular inert gas behaviour, mainly because it represents the connection between helium and fission gas in the model as it appears in the trapping rates and in the thermal re-solution rate (Table 1). Following the assumptions of [7,8,18], clusters are made of only gas atoms. and vacancy absorption at the clusters is neglected. An additional and relevant assumption is that bubbles are spherical. Consequently, the radius of intra-granular bubbles (m) is obtained as:

$$R_{ig} = \frac{3}{4\pi}V_{ig}^{1/3} = \frac{3}{4\pi}(n_{He}V_{He} + n_{FG}V_{FG})^{1/3} \quad (3)$$

where n is the number of atoms per intra-granular bubbles (at

[1] Helium, xenon, and krypton are the inert gases covered by the study, without loss of generality. Xenon is assumed to be representative of the inert fission gases. The treatment can be naturally extended to consider explicitly more inert gas species.

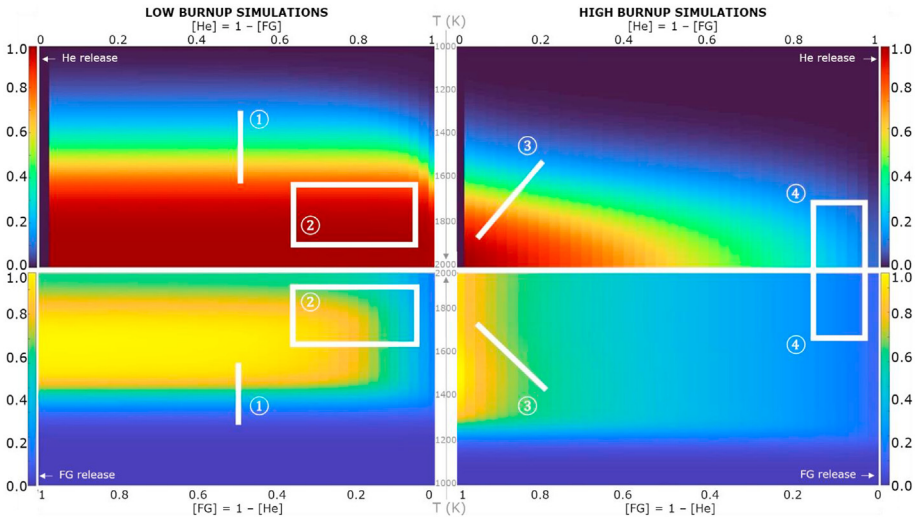

Fig. 1. Mirrored view of low and high burn-up simulations. The colour bars on the external sides indicate the quantity of intra-granular release, normalized to its maximum value. The vertical axis in the middle (specular in the lower and upper part of the map) corresponds to the temperature. The two upper quarters collect the results in terms of helium release, whereas the lower quarters collect fission gas release results. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

bubble^{-1}). V_{He} is the atomic volume of helium and corresponds to 7.8 10^{-30} m^3 [7,29]. The fission gas atomic volume in bubbles is 4.09 10^{-29} m^3, equal to the volume of a Schottky defect [8,30].

3. Showcase of results and sensitivity analysis

The model described in Section 2 is applied to the simulation of several annealing scenarios with the aim of accelerating the design and realization of new annealing experiments on inert gases. We investigate different ranges of temperature and gas composition of the cocktail to identify a region with relevant interaction between helium and fission gas within the fuel grain. The results could reduce the time and the resources needed for the realization of new experiments because they allow suggesting the most suitable starting points to investigate the behaviour of inert gases. The assumptions made for the set-up of the simulations are consistent with previous experiments in annealing conditions [17]. The temperature histories are characterized by a heat up step (from 300 K up to the final temperature in 1 h), followed by a holding at the annealing temperature for 20 h. The annealing temperatures vary from 1000 K to 2000 K (in steps of 10 K).

For the sake of showcasing the methodology, the initial quantity of fission gases within the fuel grain is calculated starting from two cases of irradiation, respectively at low and high burnup. The first case considers a period of irradiation of one week (around 0.24 GWd/t$_U$), the second case accounts for an irradiation that lasts 8 years (around 100 GWd/t$_U$). The mass of fission gas M_{FG} (g) is calculated directly from the burnup under the assumption that all the gas produced is stored in fuel grains. The mass of helium M_{He} (g) is derived as follows:

$$M_{He} = \frac{[He]}{100 - [He]} M_{FG} \quad (4)$$

where [He] corresponds to the weight percentage of helium in the grain. The use of the weight percentage instead of atomic percentage is convenient since it allows considering the effects related to the different dimensions of the atoms.[2]

The figure of merit measured up to the end of the annealing plateau is the so-called intra-granular fractional release, i.e., the fraction of either helium or fission gas leaving the fuel grain.

3.1. Recommendations

The resulting non-uniform maps (Fig. 1) clearly highlight that the interaction between helium and fission gas is to be expected, even in annealing conditions. In this regard, the simulations are intended to guide the design of future experiments on inert gases providing an extensive analysis on their behaviour and focusing the effort on the most interesting regions of study. Fig. 1 collects the results from the simulation matrix (for a total of around 4000 SCIANTIX simulations per burnup case). We highlight in white four regions which could be of interest, to investigate interactions between helium and fission gas, at low and high burn-up[3]

Besides these proposals, the construction of this type of maps highlights that, in relevant temperature/cocktail combinations, wide regions are unattractive to be investigated experimentally (e.g., virtually uniform results) whereas in specific conditions the interaction between helium and fission gas is expected to be strong and non-linear. Clearly, gathering experimental knowledge in these conditions is a much more effective strategy to assess the physics-based model parameters compared to uniformly (or randomly) exploring the full range of temperature/cocktails combinations. It is important to clarify that the model herein proposed as a guidance for the design of experimental campaigns is not fully validated. Only the extremal values of the cocktail composition are validated, i.e., [He] = 100% [7] and [FG] = 100% [22,23], in a wide range of temperature conditions. Therefore, even if the proposed model is physics-based and hence applicable outside of its strict validation range, one should use caution in doing so. For this reason, the proposed methodology is corroborated by a sensitivity analysis (in

[3] The proposed ranges of cocktail composition//annealing temperature are to be intended as examples. Other ranges may be of higher interest depending, e.g., on the available samples or on the available experimental capabilities.

[2] Using atom percentage is formally equivalent, but weight percentage ensures clearer results by providing a wider spread of the interesting phenomena in [0,100] range, whereas the use of atomic percentage compresses the results in a few percentages range.

Table 2
Scaling factors for the Pareto analysis.

Symbol	Description	Range	Reference
D_{He}	Helium diffusion coefficient	[0.1; 10]	[31]
D_{FG}	Fission gas diffusion coefficient	[0.1; 10]	[32,33]
k_H	Henry's constant	[10^{-3}; 10^{3}]	[27]
Z	Compressibility factor	[0.1; 10]	[29]

the following subsection) and should be intended as an iterative procedure, progressively incorporating in the model the information gathered by experiments. Moreover, the proposed methodology is intended as complementary to the available experimental knowledge (e.g., [20]), which should be considered in pair to the maps presented in Fig. 1.

3.2. Sensitivity analysis

In the attempt of supporting the recommendations provided, we performed a sensitivity analysis using the Pareto method. The analysis quantifies the impact of different model parameters on the model results, i.e., on the temperature/cocktail maps. The goal is again to focus the effort of future research studies on those parameters whose improvement has the priority in the enhancement of the model predictive capabilities. Considering the limited number of physical processes involved in the annealing simulations, four parameters are deemed the most representative of the model behaviour in annealing conditions, namely: helium diffusivity (D_{He}), fission gas diffusivity (D_{FG}), Henry's constant (k_H), and the compressibility factor (Z). Table 2 lists these parameters with their considered range of variation, based on published values [27,29,31–33]. The metric of comparison reported in the Pareto charts corresponds to the variation in the gas fractional release due to variation of each model parameter, averaged along the range of considered temperatures/compositions.

At low burn-up (Fig. 2) the diffusion coefficient has the major impact on the model both for helium and fission gas. This result arises from the consideration that the diffusivity is included in the mathematical description of all the processes involved in the simulations (Table 1). As for helium release, the second main model parameter is Henry's constant, governing the process of helium thermal re-solution. Its influence on the model is ascribed to the high variation range considered, based on its sensibility to temperature and on the scarcity of experimental data characterising it. It is pointed out that the impact of Henry's constant on the release of fission gases is not completely negligible, as the behaviour of fission gases is indirectly influenced by the process of helium thermal re-solution through the variation of the bubble radius (Eq. (3)). At high burn-up (Fig. 3), the Pareto chart of helium shows that Henry's constant has the main impact on the model results. This can be explained by considering that the higher mass of helium within the fuel grain (Eq. (4)) compared to the case at low burn-up and the fact that at the beginning of the simulation most helium atoms are trapped within intra-granular bubbles. Thus, the relative impact of the Henry's constant is expected to be increasing with burn-up. The compressibility factor Z has almost negligible impact

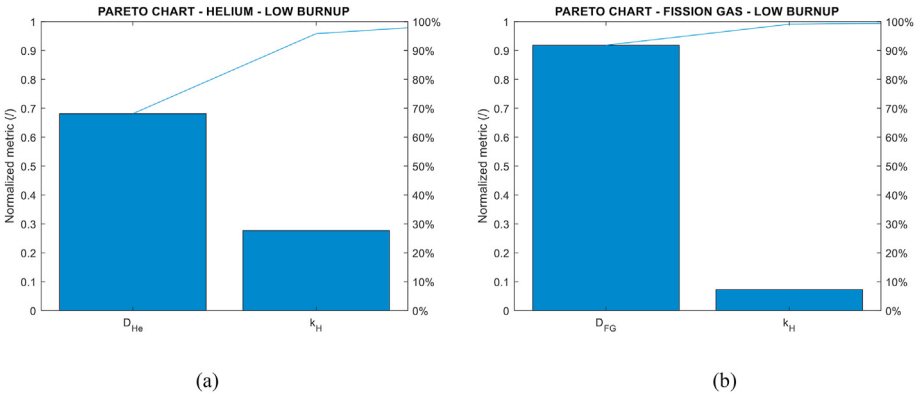

Fig. 2. Pareto charts at low burn-up, (a) focused on the impact on helium release, and (b) focused on the impact on fission gas release, respectively. Parameters with lower relative impact below 1% are not reported for the sake of clarity.

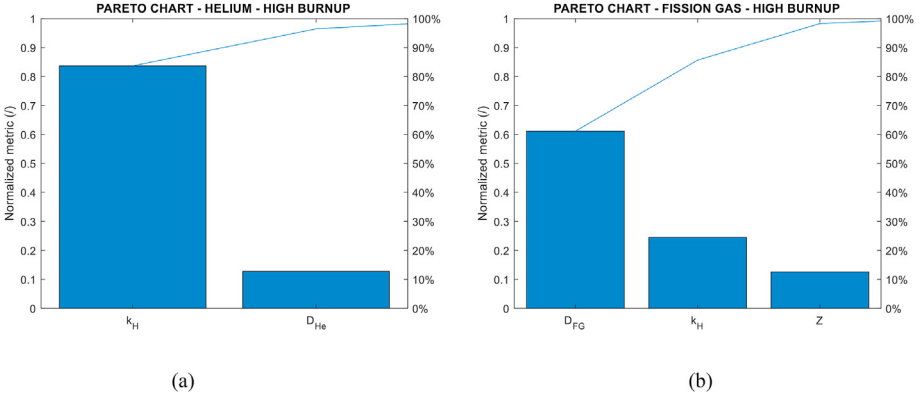

Fig. 3. Pareto charts at high burn-up, (a) focused on the impact on helium release, and (b) focused on the impact on fission gas release, respectively. Parameters with lower relative impact below 1% are not reported for the sake of clarity.

on the model in both the burn-up cases, at least in the temperature/cocktail conditions considered in this analysis. It follows that the choice of the equation of state (directly related to Z) is not the main concern for the formulation of the thermal re-solution and it has not the priority in the future development of the model.

4. Conclusions

In this work, we proposed a methodology to support the future design of experiments investigating the combined intra-granular behaviour of helium and fission gas. The first result is the development of a physics-based model which extends current inert gas behaviour models to consider the interaction of helium and fission gas within intra-granular bubbles. The model is implemented in SCIANTIX and applied in annealing conditions to explore the space of potential experimental conditions of interest.

The simulations covered the full range of helium/fission gas cocktail compositions and annealing temperatures from 1000 K to 2000 K. Two conditions have been showcased, i.e., a low burnup set-up representative of the first weeks of irradiation, and a high burnup set-up, representing highly irradiated fuel. The result of this wide simulation matrix, corroborated by a sensitivity analysis on the main model parameters, allowed to conclude that:

- The interaction between helium and fission gas is potentially relevant, with highly non-linear impact on both helium and fission gas release, at both low and high burnup.
- On the other hand, in several combinations of cocktail composition/temperature, the interaction is not expected to be strong, and thus any physics-based modelling a priori knowledge can be useful to focus experimental activities.
- Despite the coupling between helium and fission gas is connected directly to the co-presence of these species within intra-granular bubbles, the impact of the compressibility factor on their release is minimal compared to the impact of trapping and thermal re-solution of helium, at least in the range of conditions explored in this analysis. These phenomena govern the relative quantity of helium and fission gas blocked in intra-granular bubbles and therefore not available for diffusion towards the grain boundaries.

The iterative application of this methodology in synergy with an experimental campaign can provide valuable information to focus the experimental effort, with a more efficient use of resources. On the other hand, the results of such designed experiments are going to be effective in providing feedback to the proposed physics-based model, both in terms of model development and validation.

The presented analysis would benefit from several model developments, which are currently being targeted. Among the most impactful there is the treatment of grain growth (affecting release at high temperatures through the mechanism of grain boundary sweeping) and the description of the evolution of the grain-size distribution. Moreover, taking into considerations the inter-granular behaviour and/or bubble mobility may be of interest, depending on the targeted experimental conditions.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this technical note.

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