



Modelling intergranular fuel swelling in severe accidents

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Received 2 February 1999; accepted 15 June 1999

Abstract

Fuel swelling during severe accidents in PWRs is considered to be insignificant for both core degradation behaviour and fission gas release. For this reason it is ignored by the major system codes, while the complementary process of fission gas release is treated as a simple diffusion from a sphere. Recent experiments showed that fuel swelling might accelerate core degradation, and the retention of fission gases might lead to burst release in later accident phases. Modelling of swelling has been confined to full mechanistic bubble behaviour modelling (microscopic modelling), which is not suitable for the major plant codes because of its high CPU consumption. In this paper a simple analytic model based on macroscopic observations will be presented. This model uses gas diffusion from a spherical grain model for the gas atom flux into the grain boundaries, and vacancy diffusion from the pellet surface model for the vacancy flux to the grain boundaries. The total vacancy volume and the gas atom number are coupled by the Xe-equation of state. The gas release or fuel swelling is then pressure controlled. The model gave good results for both release and swelling when compared with experimental observations. This made clear that the application of the gas diffusion model alone is not enough to describe the fission product release. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

Fuel swelling is the process during which the fuel expands in volume due to fission products trapped in it. Fission products are created in the fuel matrix during normal operation. They can be solids or gases. During a severe accident swelling is caused by fission gases, such as Xe, Kr, Cs etc. and it can reach more than 100% of the original fuel pellet size.

Fuel swelling, although very important for fast breeder reactors (LMFBR), until recently was not considered a significant phenomenon for PWRs. In a PWR the concentration of the fission product gas is not so high as in an LMFBR. The temperature escalation during a PWR severe accident, unlike that in an LMFBR, proceeds slowly enough to allow the fission gases to be released from the fuel with only negligible swelling. In low pressure sequences the fuel rod cladding fails at temperatures of only 750–850°C and the depressurisation of the rod enhances the fission gas release. This was the established opinion about swelling, until

experiments such as ST-1 [3], FLHT-5 [4] or even PHEBUS FPT-1 [2,23] showed that there can be significant fuel swelling under PWR severe accident conditions as well, when temperatures go above 2000°C.

Swelling affects the fuel performance in two ways [5,7]:

- (a) It causes fuel-cladding contact. This has two effects:
 - (i) Earlier clad failure because of local stresses internally or even clad melting due to the higher fuel temperature.
 - (ii) Fuel liquefaction and dislocation because of the U–Zr contact. The first is relevant if the clad has not already failed. In a typical low pressure PWR accident sequence this is not probable because clad failure is an early phenomenon. In a high pressure sequence fuel-cladding contact is more probable although the high pressure will not favour bubble growth and thus swelling. The second is of great interest as it greatly affects the late phase of the core degradation such as debris-bed and molten pool formation.
- (b) It changes the fuel physical properties such as thermal conductivity or density [6]. This results in

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the worsening of the heat exchange between fuel and its environment, which leads to earlier fuel liquefaction, which again affects the late phase of the core degradation. On the other hand swelled fuel is harder to quench.

Moreover it is decisive for the fission gas release. The greater the swelling the greater the gas that remains in the fuel, i.e. the lower the fission product release. In later accident stages, when melting occurs, it may lead to burst release with significant consequences for the source term.

2. Phenomenology of swelling

Fission gas atoms are dispersed in the fuel solid matrix. The longer the normal operation period the greater the amount of those gases in the fuel. Defects, such as vacancies and interstitials are also produced during normal operation [5,8].

During a severe accident fuel temperatures increase in short time periods (accident dependent). Gas atoms become mobile and migrate towards the grain boundaries. On this motion they are trapped by vacancies, dislocations and other defects and they form bubbles [5,8,10,16]. One can distinguish between intragranular and intergranular gas.

Intragranular gas is the gas residing in the fuel grains. This gas may be dispersed in its molecular form or accumulated in bubbles. Intragranular bubbles are generally small in the first accident stages and contribute to swelling only at very high burn-ups as long as fuel is in its solid state [12]. This situation changes when the fuel starts to liquefy. The enlargement of tiny bubbles may lead then to frothing.

Conversely gas residing in grain boundaries (intergranular) finds more space to expand. Due to gas diffusion from inside the grain to grain boundaries, the gas amount increases leading to larger and larger bubbles, which interconnect forming pathways for fission product release. It has been observed [16,12,8] that the fission product release or swelling depends on:

The heat-up rate: A slow heat-up will lead to slow and gradual fission product release without significant swelling, while a fast one may lead to fast release from the grains and an accumulation of the gas in the grain boundaries as the necessary pathways are not yet formed to allow release outside the pellet. In this case swelling will occur. Bagger et al. [6] observed a dynamic behaviour in the Xe release that depended on the temperature escalation. In order to keep the model of simple gas diffusion from a sphere they had to define two different diffusion coefficients, one for fast and one for slow escalations.

Gas amount: The more the gas in the fuel, i.e., the higher the burn-up, the more the swelling will be [12].

System pressure during the accident: The higher the system pressure the more gradual the fission product release. Low system pressure may lead to burst release if there is a lot of gas in the fuel [11].

Some codes have been developed to treat mechanistically this problem. They model the intra-and intergranular bubble behaviour such as bubble nucleation, bubble growth, bubble migration etc. Such codes have not been incorporated in large system codes such as MELCOR, ICARE2 etc., as their computational time is too high and would slow up the anyhow CPU-costly calculation. Thus swelling is ignored in system codes whereas the swelling-connected phenomenon of fission product release is modelled independently either as a simple correlation or by using the gas diffusion from a spherical grain model (Booth model) [19]. The latter used on its own seems to give reliable results in slow transients, whereas in case of sudden temperature increase it will overestimate the release as the grain boundaries are not considered, and thus it will underestimate it later on in the transient. Attempts have been made to overcome this problem by introducing the diffusion distance instead of the grain radius; but the choice of this parameter seems rather difficult and it cannot be defined a priori.

3. A different approach

Although vacancies and gas atoms are also present in the fuel during normal operation, swelling does not occur. It is significant only during heat-up in abnormal conditions. Due to the higher temperature the gas needs to expand. It moves towards vacancy rich regions such as pores, grain boundaries or dislocations and absorbs all the vacancies around it creating bubbles. The gas overpressure in the fuel can be reduced only by gas volume increase. The fuel matrix starts to expand by increasing the rod diameter. The volume increase is equivalent to the free volume flowing into the fuel in form of vacancies. Vacancy migration occurs:

- (a) from the pellet rim (cold region with low gas concentration) towards the pellet interior (hot region with high gas concentration),
- (b) from the grain boundaries towards the grain centres.

The first process is faster as both vacancy and gas diffuse easier at the grain boundaries (surface diffusion) than in the grains themselves (volume diffusion), and the first to occur, leading to intergranular swelling. Vacancies that migrate through the grain boundaries are trapped by the gas atoms, which also migrate there creating continuously growing bubbles. After reaching a certain size the

bubbles interconnect creating pathways for the fission gas release [16]. Gas acts as a perfect sink for vacancies as it expands. The pellet rim is a perfect source for vacancies as the fuel matrix atoms can move easily there. Vacancies then diffuse due to the concentration gradient towards their sink in the pellet. The interconnection of the intergranular bubbles will start from the hotter pellet inside, as the gas release from the grains is higher there and will expand towards the colder pellet rim. The pellet rim is the last barrier for the fission product release. Just before this barrier fails it can be assumed that the whole gas is interconnected and occupies a single volume. At that stage the maximal grain boundary swelling has been exceeded.

Grain boundary swelling occurs in the first stages of the temperature escalation and only if the fuel is in solid state. Kashibe et al. [12] showed clearly in their experiments with different burn-ups that the grain region next to the grain boundaries totally deforms due to big bubbles, whilst in the grain itself bubbles remain tiny. This shows that vacancies also diffuse from the grain boundaries towards the grain centre, as there the highest gas concentration occurs. Evans [17] tried to model this as a region of high vacancy diffusion and to treat in this way both release and swelling.

When the fuel starts to change its state, i.e. from solid to liquid, gas diffusion and bubble migration towards the pellet rim become the dominant processes. On the way out bubbles collide with gas atoms and become larger. This phenomenon is referred to as frothing.

The phenomenon of vacancy flow towards the interior of the fuel occurs fast in the first stages of the swelling. It is difficult to observe in the standard annealing tests as in those the fuel is kept for long periods at a high temperature giving enough time to the pore interconnection and thus to fission gas release. Intergranular bubbles that reach the pellet rim also transport vacancies to the pellet rim; thus the slow fission product release in annealing tests is coupled with a vacancy flow towards the pellet rim. The slower phenomenon of vacancies flowing from the grain surface towards grain centre has however been observed in the annealing tests [12–14].

4. Model

The aim was to treat the above mentioned phenomena by a simple analytical model. Fuel is packed in pellets pressed together in rods of a height of 4 m. The radius of the pellets is not more than a few millimetres. It is obvious that swelling in the axial direction is negligible and that radially the one-dimensional model in cylindrical coordinates is quite sufficient. We propose a macroscopic model, where the release and swelling of the whole pellet is treated simultaneously. In this first

development stage only intergranular swelling is considered. The model operates in four steps:

- gas diffusion from the spherical grains towards the grain boundaries (Booth model [19]),
- vacancy diffusion from the cylinder surface towards the cylinder centre,
- coupling of gas atoms and vacancies by the equation of state of Xe assuming that all the gas is interconnected and thus forms a single volume,
- comparison of the gas pressure to the system pressure as criterion for release or swelling.

4.1. Gas release to grain boundaries

As long as the fuel is in solid state gas will be released by diffusion to the grain boundaries. The grains are assumed to be spherical and bubbles, dislocations and other kind of defects are all included in the diffusion coefficient. It can be described by the Booth model, where the release fraction F is given [18,19] by

$$F = \begin{cases} 6 \left(\frac{D_g t}{\pi a^2} \right)^{1/2} - 3 \frac{D_g t}{a^2} & \text{for } \frac{\pi^2 D_g t}{a^2} \leq 1, \\ 1 - \frac{6}{\pi^2} e^{-\pi^2 D_g t / a^2} & \text{for } \frac{\pi^2 D_g t}{a^2} \geq 1, \end{cases} \quad (1)$$

where D_g is the gas diffusion coefficient in m^2/s , a the grain radius in metres and t the time in seconds.

The released gas will remain in the grain boundaries forming bubbles with the existing vacancies. The existing vacancies in the fuel are not sufficient for swelling to occur. Vacancies start to migrate from the pellet rim. The more gas there is, the more vacancies are required.

4.2. Vacancy diffusion from the pellet rim

During the temperature escalation a large number of vacancies will migrate from the pellet rim (cold) towards the pellet centre (hot) driven by the absence of vacancies due to the gas expansion. We assume vacancy diffusion in the r -direction of the cylindrical coordination system. A pellet can be approximated with a cylinder of radius R .

The diffusion equation in cylindrical coordinates is given by

$$\frac{dC(r, t)}{dt} = \frac{1}{r} D_v \frac{\partial}{\partial r} \left(r \frac{\partial C(r, t)}{\partial r} \right), \quad (2)$$

where $C(r, t)$ is the vacancy concentration in m^{-3} and D_v vacancy diffusion coefficient in m^2/s ,

The solution of Eq. (2) is given in [18] for the initial and boundary conditions:

$$C(r, 0) = C_1, \quad (3)$$

$$C(R, t) = C_0 \quad \text{and} \quad C(0, t) = \text{finite}, \quad (4)$$

where R is the radius of the pellet in metres and C_0 and C_1 are the boundary and the initial condition respectively, with

$$\frac{M_t}{M_\infty} = \begin{cases} 1 - \sum_{n=1}^{\infty} \frac{4}{R^2 \alpha_n^2} \exp(-D_v \alpha_n^2 t) & \text{if } \frac{D_v t}{R^2} \geq 1, \\ \frac{4}{\sqrt{\pi}} \sqrt{\frac{D_v t}{R^2}} - \frac{D_v t}{R^2} - \frac{1}{3\sqrt{\pi}} \sqrt{\frac{D_v t}{R^2}} & \text{if } \frac{D_v t}{R^2} < 1, \end{cases} \quad (5)$$

where α_n are the solutions of $J_0(R\alpha_n) = 0$ and J_0 is the Bessel function zeroth order, M_t the diffusing amount of vacancies at time t and M_∞ the diffused vacancies after infinite time.

The equilibrium vacancy number is taken for M_∞ , since we assume that at the defect-free pellet surface thermal equilibrium is immediately established. Thus

$$M_\infty = \Omega C_v^{\text{eq}} = \exp(-e_f/kT), \quad (6)$$

where e_f is the activation energy for vacancy formation in eV, k the Boltzmann constant and T the temperature in Kelvin. The driving force for the vacancy diffusion is the gas excess-pressure.

4.3. Equation of state

These vacancies are absorbed by the gas forming continuously growing bubbles, until those interconnect and gas release can start. We assume that other vacancy sources such as dislocations etc. contribute only marginally to the total volume, while the initial porosity is treated separately. For the model's last step we assume that the gas is already interconnected, i.e. the whole gas residing in the grain boundaries forms a single large volume. This single volume will not stop absorbing vacancies and growing, causing swelling, until its pressure has fallen to the outside pellet (system) pressure P . Then release starts.

From Eq. (1) we have the gas atoms N_t released from each grain in each time step. From Eq. (5) we have M_t which represents the available space for the gas expansion due to the diffusing vacancies. When using the van der Waals equation of state for Xe we have

$$P_g(V_v - N_t w) = N_t kT, \quad (7)$$

where P_g is the gas pressure in N/m² and

$$\text{if } \begin{cases} P_g \leq P & \Rightarrow \text{release,} \\ P_g > P & \Rightarrow \text{swelling.} \end{cases} \quad (8)$$

$V_v = M_t \times \Omega$ is the vacancy volume, where $\Omega = 4.09 \times 10^{-29}$ m³ per atom, U the atomic volume, $N_t = F \times N_x$ where N_x is the total gas atoms number in the fuel given by the fuel burn-up and w the van der Waals constant for Xe given as 8.5×10^{-29} m³.

Depending on the number of vacancies and the amount of fission gas atoms released, Eq. (8) will either allow release from the fuel or swelling; but not both. A

certain amount of vacancies will be released with the gas. According to Olander [5] each gas atom requires 27 vacancies. This means that with each released gas atom 27 vacancies are released.

Only the presence of gas under excess-pressure can drive the vacancy diffusion mechanism. If there is an initial porosity, i.e. free volume for the gas already at the beginning of the heat-up, gas release will not occur although the gas pressure may be still smaller than the system pressure. Once the first excess-pressure is built up the vacancy diffusion starts and Eq. (8) can be applied. The initial porosity is given by the fuel manufacturer in per cent of fuel volume. This volume is taken into account for the fission product expansion.

In this way a relatively simple solution can be obtained and the experimental observations are satisfied, particularly:

- No sudden release occurs in case of sudden temperature escalation, since the gas cannot release if the grain boundaries are not interconnected, but burst release may also occur if a lot of gas has accumulated in the fuel.
- The effect of the gas amount is taken into consideration. Thus small gas amounts are released more easily than large amounts, which lead to swelling.
- The higher the outer pressure the 'smoother' the release and the less the swelling.

The model does not apply when the temperature is close to melting point or above it. The effects of mechanical stresses, dislocations or cracks are not considered. Such phenomena act as vacancy sources and would increase the swelling.

The swelling is then given in per cent by

$$S = \frac{V_v}{V} \times 100, \quad (9)$$

where V_v is the volume of the vacancies that have penetrated the pellet, subtracting those that have left the pellet with the gas, V the fuel volume considered.

5. Results

In the case of a severe accident a temperature history is provided and we want to estimate the release and swelling over the temperature escalation period. Diffusion coefficients D_g and D_v and vacancy equilibrium number M_∞ depend on the temperature. Thus Eq. (1) becomes

$$F = \begin{cases} 6 \left(\frac{\sum_i D_g^i \Delta t_i}{\pi a^2} \right)^{1/2} - 3 \frac{\sum_i D_g^i \Delta t_i}{a^2} & \text{for } \frac{\pi^2 \sum_i D_g^i \Delta t_i}{a^2} \leq 1, \\ 1 - \frac{6}{\pi^2} e^{-\pi^2 \sum_i D_g^i \Delta t_i / a^2} & \text{for } \frac{\pi^2 \sum_i D_g^i \Delta t_i}{a^2} \geq 1, \end{cases}$$

and Eq. (5)

$$M_t = \begin{cases} \sum_i \left[1 - \sum_{n=1}^{\infty} \frac{4}{R^2 \alpha_n^2} \exp(-D_v^i \alpha_n^2 \Delta t_i) \right] M_{\infty}^i & \text{if } \frac{D_v^i \Delta t_i}{R^2} \geq 1, \\ \sum_i \left[\frac{4}{\sqrt{\pi}} \sqrt{\frac{D_v^i \Delta t_i}{R^2}} - \frac{D_v^i \Delta t_i}{R^2} - \frac{1}{3\sqrt{\pi}} \sqrt{\frac{D_v^i \Delta t_i}{R^2}} \right] M_{\infty}^i & \text{if } \frac{D_v^i \Delta t_i}{R^2} < 1, \end{cases}$$

where

$$D_g^i = 30.4 \times 10^{-10} \exp(-35400/T_i),$$

$$D_v^i = 57 \exp(-54400/T_i),$$

the vacancy formation energy was taken $e_f = 1.0$ eV [5]. D_v is the standard UO_2 -surface diffusivity. The gas diffusion coefficient is the one used by the ELSA [20] code, which has recently been implemented in the ICARE2 [21] code and was recommended based on the VERCORs tests. The UO_2 surface diffusivity is the one used by the code LAKU [22]. Similar values are given elsewhere in the literature such as by Evans [13].

Three cases will be presented here, one of low burn-up fuel and two of moderate burn-up fuel. The difference between the latter two will consist in the rate of the temperature escalation. The test volume is a pellet of 1mm height and 4mm radius. We assume a low pressure case with system pressure $P = 2.2$ bar. The fuel characteristics and the temperature escalation correspond to the values of the two PHEBUS tests FPT-0 and FPT-1.

5.1. Low burn-up fuel

Two cases will be shown with the same temperature escalation. In the first the initial porosity of 4% will be taken into account. In the second the fuel will be assumed re-densified with 0% initial porosity. The typical gas amount (Xe, Kr, Cs) of such a fuel in the test volume is $N_x = 8 \times 10^{-5}$ g or 10^{16} gas atoms, values as for FPT-0 [1,23]. The temperature escalation corresponds to calculated values of the test for the time period from 1000 to 18000 s.

Case A. In Fig. 1 is shown the temperature escalation, the gas release according to Eq. (1) and the gas release according to the criterion of Eq. (8). In the second part of Fig. 1 the swelling is shown.

In this case neither swelling nor release occur until a certain time is reached. The reason is the initial porosity which retains a great amount of the gas before release. Swelling does not occur as there is no need for vacancy diffusion. Moreover when release starts the fuel becomes denser as many vacancies leave the fuel with the gas atoms. Thus swelling is -0.3% i.e. shrinkage of 0.3%.

Case B. Since in this case there is no porosity the gas released in the grain boundaries causes swelling but a very small one, as the gas amount is low in the initial phase of the temperature escalation. Later the gas release follows the Booth model. The sudden temperature increase at about 1000 s causes high release to the grain boundaries, while not enough vacancies have entered the pellet. The gas pressure according to Eq. (7) rises and swelling occurs. The small gas amount causes a swelling of only 0.02% (see Fig. 2).

5.2. Moderate burn-up fuel with slow transient

We consider the same cylinder but now the amount of the same gases is $N_x \sim 8 \times 10^{-3}$ g or 10^{18} gas atoms. Here the initial porosity is 6%, values as in FPT-1 [2,23]. Again the temperature escalation corresponds to simulated values for the time period of 9000–17200 s. In Fig. 3 one can see the temperature, the release and the swelling. Again swelling and release do not start until the available volume of the porosity is used up. Calculations with 0% porosity changed the result only negligibly as in this case it is the high gas amount that governs the fuel behaviour and not the initial porosity. In the same way as previously swelling occurs during the temperature escalation phase. In this case the released gas is much more and thus also the swelling which

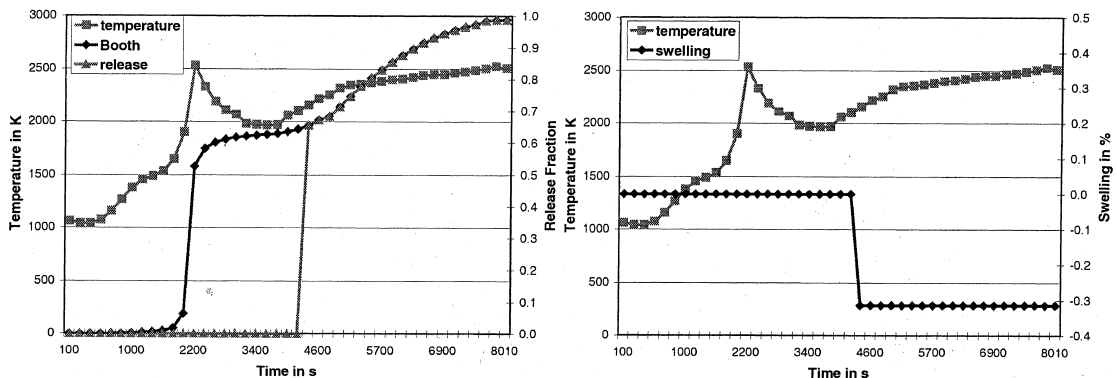


Fig. 1. Release and swelling in the low burn-up case with 4% porosity.

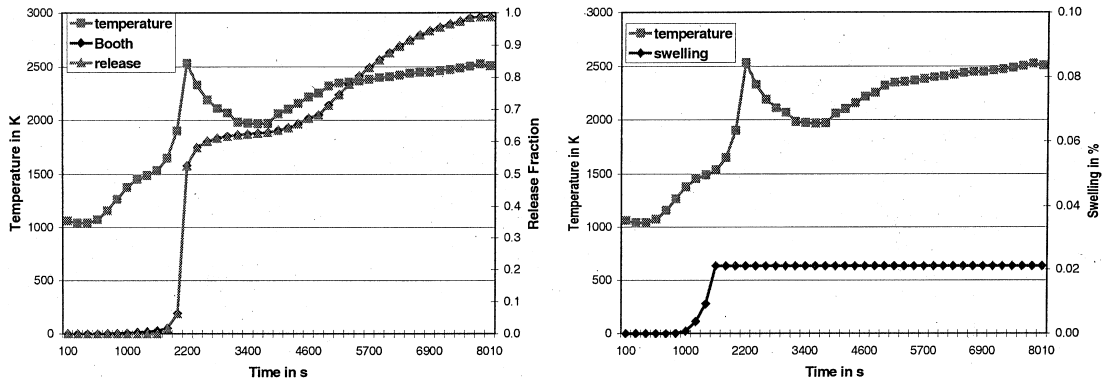


Fig. 2. Release and swelling in the low burn-up case with 0% porosity.

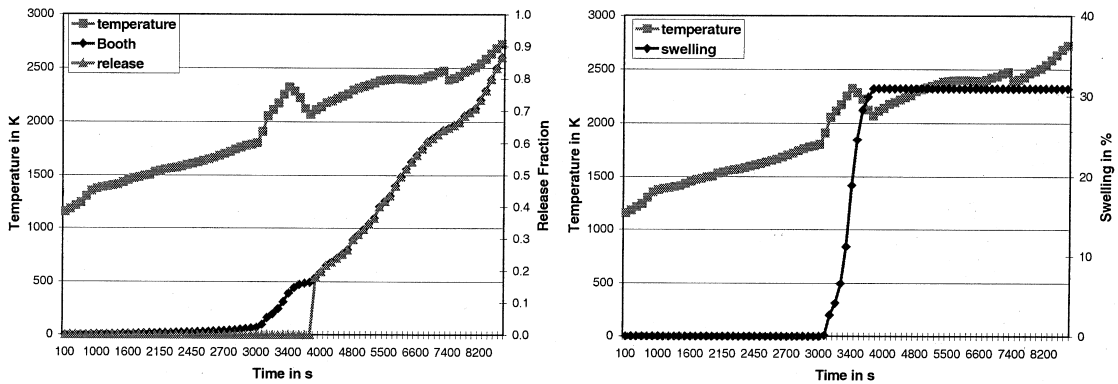


Fig. 3. Release and swelling in the moderate burn-up case slow transient.

reaches ~31%. This value is close to the observed value of about 25%.

5.3. Moderate burn-up fuel with fast transient

The temperature escalation is now as in Fig. 4. This escalation corresponds to escalations observed in

FLHT-5 [4]. In this very fast escalation the temperature rises from 100 to 3000 K in a few seconds. One can see the difference between the releases by using only the Booth model or the new approach. Intergranular swelling now reaches 71% (Fig. 4) as the escalation is much faster. In FLHT-5 swelling with frothing was observed, which reached partly 400%.

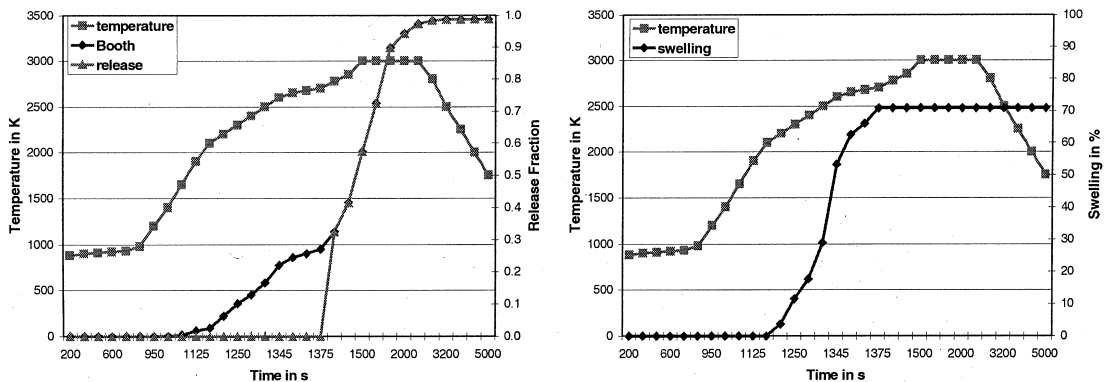


Fig. 4. Release and swelling in the moderate burn-up case with fast transient.

5.4. Comparison with the LAKU code

The LAKU code was run for the same test cases and under the same conditions as used in the proposed model. The model's results are here compared with results of the LAKU code. The first figure concerns the FPT-1 test, the second the FLHT-5 experiment. In both cases the LAKU code and the presented model show the same swelling dynamic. Swelling increases during the heat up period. LAKU calculates a lower swelling for the FPT-1 test. The difference is due to the abrupt swelling/release condition (Eq. (8)) used in the current model. The same occurs in FLHT (right part of Fig. 5), though there the high temperatures cause a large intragranular swelling and frothing, which are not included in the model presented here. The second part of Fig. 5 is focused on the 1000 s of the temperature escalation, i.e. from 1000 to 2000 s of the time scale used in Fig. 4.

6. Conclusions

A model for intergranular swelling was presented. This model is based on the assumption that vacancies will diffuse from the pellet rim to the pellet centre due to the concentration gradient produced by the gas' need for expansion. Thus the grain boundary gas acts as a perfect sink for vacancies. Gas diffuses to grain boundaries from the interior of the grain. The model was found treat very well the sudden temperature escalations that are the reason for swelling. It gave reasonable results for both low and moderate burn-up fuel. The difference between fast and slow temperature escalation was made clear. The 400% swelling observed at FLHT-5 could not be calculated for two reasons; firstly the intragranular swelling is not considered and secondly the melting temperature was

reached in this experiment and thus frothing occurred. This phenomenon is also not considered in the presented model.

The biggest uncertainties of the model are gas and vacancy diffusion coefficients and the vacancy formation energy. These three parameters depend strongly on the fuel burn-up and degree of fuel oxidation. A series of small separate effect experiments for the estimation of these values at different burn-up and oxidation degrees would help the performance of the models considerably. One though has to bear in mind that this model has a smaller amount of uncertainty variables in comparison to the mechanistic bubble models in which variables such as the radial fuel temperature gradient or the bubble coverage coefficient [9] can hardly be estimated.

The values of the gas and UO₂ surface diffusion coefficients are generally accepted and used in other codes while the vacancy formation coefficient at the surface is low in comparison to other suggested values, such as 1.6 eV by Rest [15]. The latter value gave unrealistic high swelling values, which suggest that the produced vacancy amount at the pellet rim was underestimated. The value of 1 eV gave good results for all the experiments considered in this study, though it has still to be justified.

The number of vacancies living the fuel with the gas was not found to play a significant role. In fact the results change only marginally when the value of 2 vacancies per atom are used instead of 27.

Work is being done in improving the release/swelling criterion (Eq. (8)). In its present form either the one or the other can occur. A 'softer' criterion, which would allow a transition period where both can occur, would be more appropriate.

As the model does not apply for melting conditions, it has to be extended to include grain swelling and frothing as both these mechanisms may affect significantly the later stages of the accident.

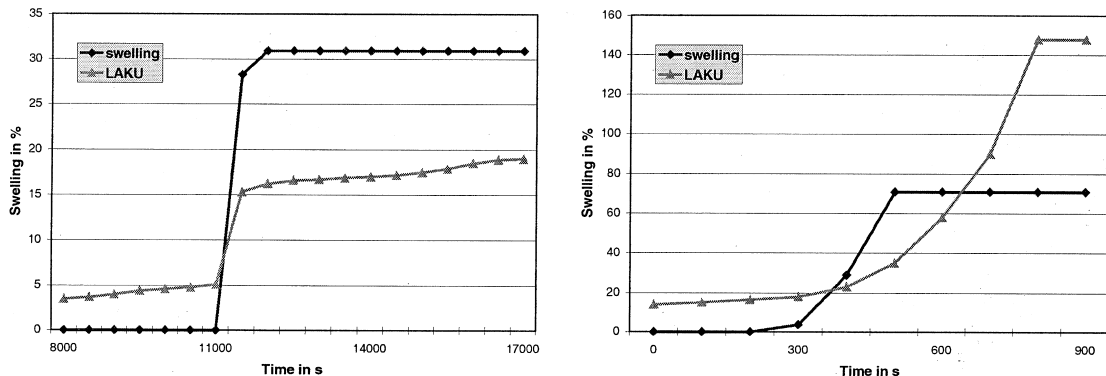


Fig. 5. Comparison between the model and the LAKU code.

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