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RECENT DEVELOPMENTS OF THE TRANSURANUS CODE WITH EMPHASIS ON HIGH BURNUP PHENOMENA

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Abstract

TRANSURANUS is a computer program for the thermal and mechanical analysis of fuel rods in nuclear reactors, which is developed at the Institute for Transuranium Elements. The code is in use in several European organisations, both in research and industry. In the paper the recent developments are summarised: the burnup degradation of the fuel's thermal conductivity as well as the effects of gadolinium on the radial power distribution and thermal conductivity. Fission gas release from the High Burnup Structure is discussed. Finally, a new numerical method is outlined that is able to treat the highly non-linear mechanical equations in transients (RIAs and LOCAs).

1. INTRODUCTION

TRANSURANUS is a computer program for the thermal and mechanical analysis of fuel rods in nuclear reactors, which is developed at the European Institute for Transuranium Elements [1]. The code is in use in several European organisations, both in research and industry and is under continuous development. In the following paper recent developments are outlined with emphasis on high burnup models.

2. THERMAL CONDUCTIVITY OF LWR FUEL AT HIGH BURNUP

In view of its relevance an extensive review of the degradation of the thermal conductivity with burnup was performed. In order to compare the various correlations in the literature, a conversion to the simplest form of the phonon term

$$l \quad \frac{1}{a_{1} \quad a_{1}\, bu \quad bT} \qquad (1)$$

was made, where l is the thermal conductivity, T is the temperature and a_{1}, a_{1} and b are parameters. Figure 1 shows that there is a very consistent picture of the coefficient a_{1} from References [2-9]. In addition, it can be seen that the results from Simfuel measurements [10-12] gave a significantly lower value, due to the fact that radiation effects such as the formation of gas bubbles are not accounted for.

Traditionally, the thermal conductivity is formulated as a term related to a specified density multiplied by a function of the local porosity. Thus the enhanced porosity in the high burnup structure (HBS) needs to be considered, resulting from the formation of a new bubble population. The level of the increase in porosity is still a matter of discussion. No real progress has been made for several years. Density measurements indicate that the measured porosity in highly irradiated samples may be affected by grain pull out leading to an overprediction of the porosity. Based on the data of one of the authors, Vennix [13], it was possible already in the years 1995-1996 to establish a clear trend by converting the porosity as a function of the radius to porosity as a function of the local burnup. Similar measurements have been made by other authors, for instance by Spino [14, 15]). The original data of Vennix are given in Figure 2. Using the TRANSURANUS burnup model, the local burnup can be calculated for each data point and the data can be correlated with the local burnup. The results are shown in Figure 3. Magnifications between 500× and 1600× were used, which give very consistent results. Above approximately 50000 MWd/tU the porosity increases linearly with the local burnup. We observe the same trend for the pore density which clearly saturates at about 100000 MWd/tU. Figure 3 a) is the basis for the correlation

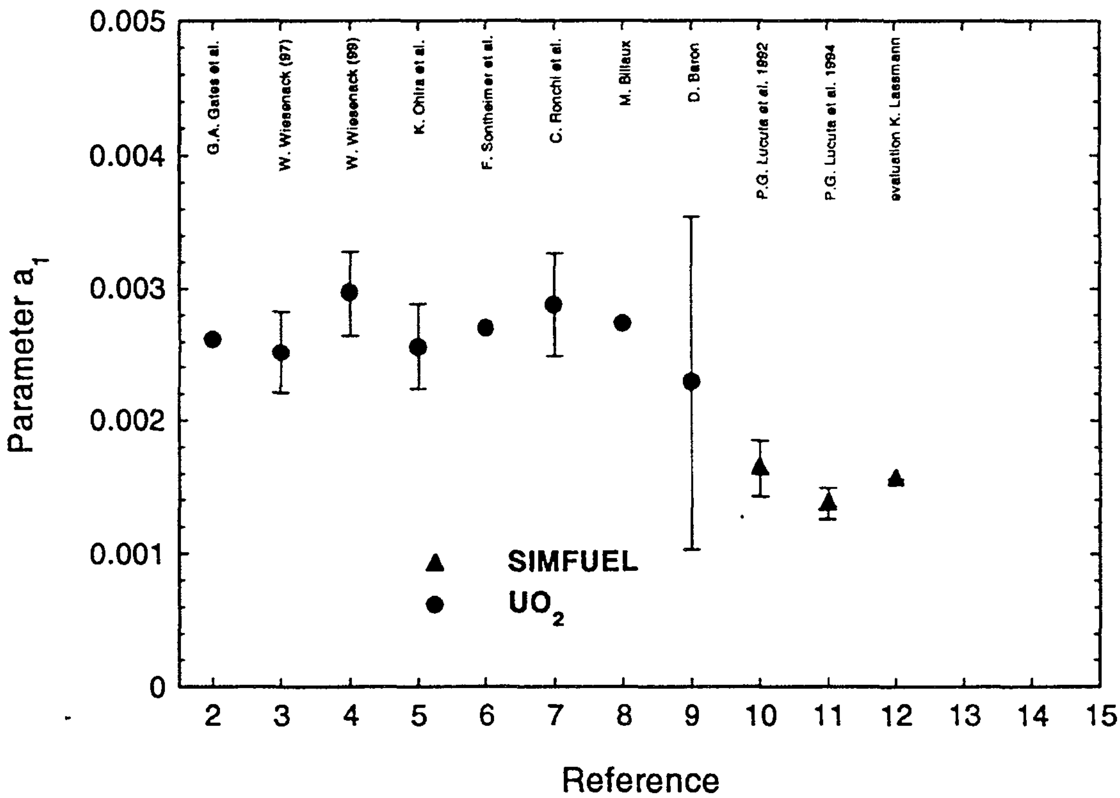


Figure 1. Value of the coefficient a1 , which describes the degradation of the thermal conductivity with burnup, according to various references. Simfuel gives a significantly lower value than irradiated UO_2.

$$\Delta P^{HBS} = 1.29 \cdot 10^{-6} \, \Delta bu \qquad \text{if } bu \geq 60000 \frac{MWd}{tU} \tag{2}$$

incorporated in the TRANSURANUS code. ΔP^{HBS} is the increase of the additionally formed porosity in the High Burnup Structure during the burnup increment Δbu. The maximum increase of this type of porosity is limited to 13%. Equation (2) has been included in Figure 3a. Also included are some first estimations derived from density measurements. Although there seems to be a good agreement, further clarification is needed.

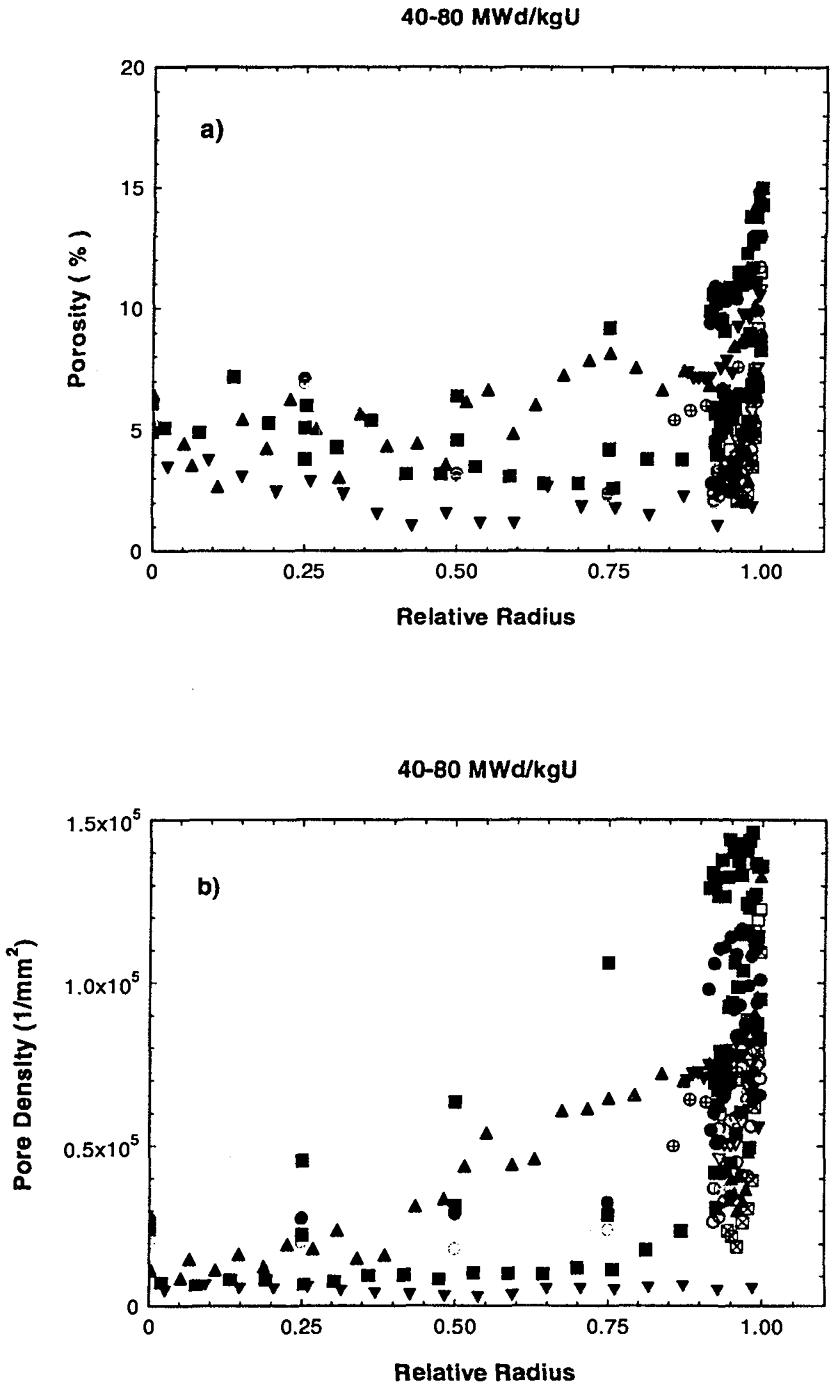
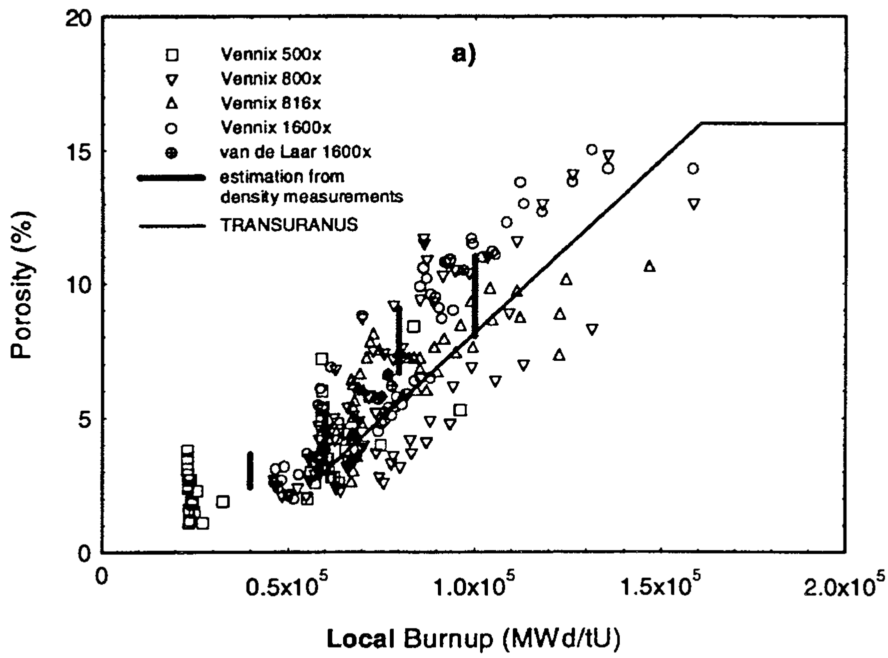


Figure 2 a). Porosity and b) pore density as a function of the radius for highly irradiated fuel according to the data of Vennix [13]. Note that these diagrams are used to investigate trends only (see Figure 3).

The trend of the porosity shown in Figure 3a was questioned by Sontheimer [16] and has not been confirmed by other authors. We consider that the reason lies in the high experimental uncertainties caused by difficulties in specimen preparation resulting in strong variations of measured porosities by different groups (and even individuals). It is interesting to note that a similar trend (although with higher porosities) as in Figure 3a results when the data of Spino are evaluated in the same way.



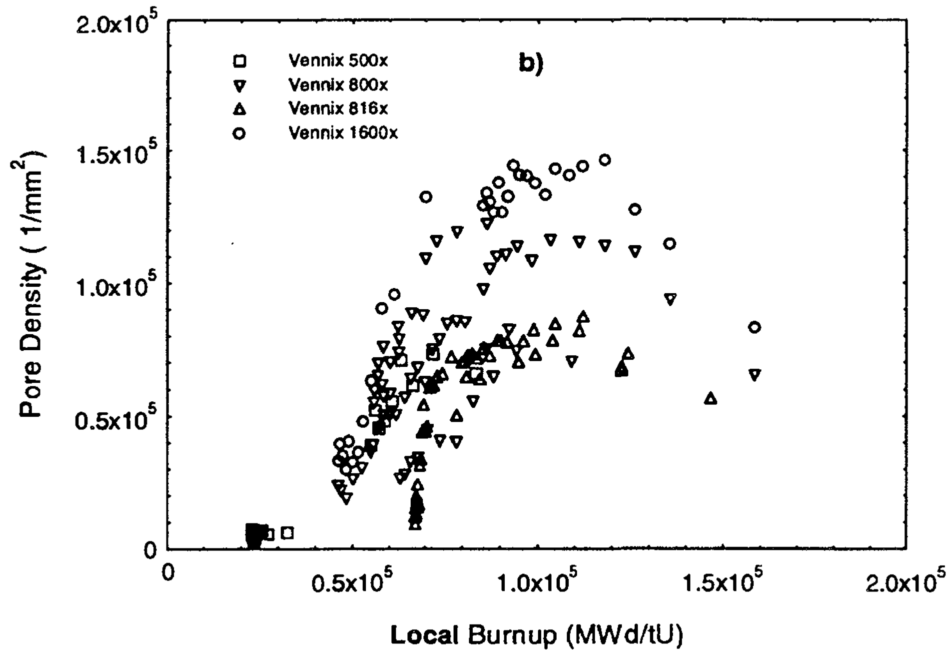


Figure 3. a) Porosity and b) pore density as a function of the local burnup for highly irradiated fuel. The data are the same as in Figure 2. Note that these diagrams are used to investigate trends only.

It is evident that the clarification of the porosity in the high burnup structure is of extreme importance since the expansion of the fuel due to the formation of porosity as well as the decrease of the thermal conductivity influence the thermal and mechanical behaviour. In spite of this importance, in none of the papers on the thermal conductivity of UO_2 at high burnup this question is addressed sufficiently.

Most correlations for the thermal conductivity from the open literature are incorporated into the TRANSURANUS code. An own correlation, developed from ITU measurements of Ronchi and Sheindlin [7], summarises our present-day knowledge. This correlation includes the porosity correlation (2) and the effect of gadolinium. Figure 4 shows the good agreement between this correlation and measured data for a wide range of conditions: UO_2 with $0 \le burnup \le 100000\ MWd/tU$ and Gd_3O_2 contents up to 19 wt% (References 7, 17, 18, 19).

3. TREATMENT OF BURNABLE ABSORBERS

The need to improve reactor performance through longer cycle lengths or improved fuel utilisation has been apparent since the beginning of commercial nuclear power generation. The fuel initial enrichment has been increased, with the consequence that the additional amount of fissile material in the core has had to be compensated for by the introduction of additional absorber material in the core [20]. This additional absorber can be introduced in the form of

- control rods
- soluble absorber (boric acid) in the coolant
- burnable absorbers inside the fuel (integral burnable absorbers).

For all BWRs integral burnable absorbers in the fuel are chosen, for PWRs the use of soluble absorber in the coolant was for many years standard. However, the increase of initial fuel enrichment cannot be indefinitely compensated for by increasing the boric acid concentration and therefore integral burnable absorbers are now considered in PWR designs also.

Two concepts of integral burnable absorbers are treated in the TRANSURANUS code:

- Gadolinia (Gd_2O_3)
- Zirconium diboride (ZrB_2).

These are discussed below.

3.1. Gadolinia (GD_2O_3)

A gadolinium content in the fuel is considered to have several significant effects on fuel performance

- It degrades the thermal conductivity of the fuel.
- It reduces the melting point.
- It produces a distorted, rapidly changing radial power profile.

Natural gadolinium consists of seven isotopes with mass numbers 152, 154, 155, 156, 157, 158 and 160. The natural percent abundances are 0.2, 2.1, 14.8, 20.6, 15.7, 24.8 and 21.8, respectively. This burnable absorber works by neutron capture of the two isotopes, ^{155}Gd and ^{157}Gd, with extremely high absorption cross-sections. The isotopes produced by this reaction, ^{156}Gd and ^{158}Gd have a small absorption cross-section and need not to be further considered.

The methodology to describe the neutron absorption of ^{155}Gd and ^{157}Gd is similar as for the standard TRANSURANUS burnup equations (for details, see Ref. [21], Eq. 3).

The difficulty in this simplified treatment is the definition of the effective absorption cross-section σ_a since the absorption cross-section is extremely high for neutron energies below 1 eV. The consequence is

- a local shift of the neutron spectrum of the thermal flux (hardening) and
- a significant spatial selfshielding of the fuel.

We have tried to fit the effective absorption cross-section in a reasonable way, i.e., the local change of the neutron spectrum is approximately taken into account. The selfshielding is treated by solving the diffusion equation

$$\nabla^2 f - \kappa^2(r)\, f = 0 \qquad (3)$$

numerically. κ is the inverse diffusion length which depends on the radius.

This gadolinium model was fitted to the average concentrations of ^{155}Gd and ^{157}Gd of Fuji et al. [22]. Figure 5 shows a reasonable agreement with the data.

Having fitted the two parameter of the TRANSURANUS Gd burnup model, the model can be applied to predict the radial ^{155}Gd and ^{157}Gd profiles at different burnups. Reference [22] reports such data, which are compared to the model predictions in Figure 6 and Figure 7. As can be seen, the results are acceptable.

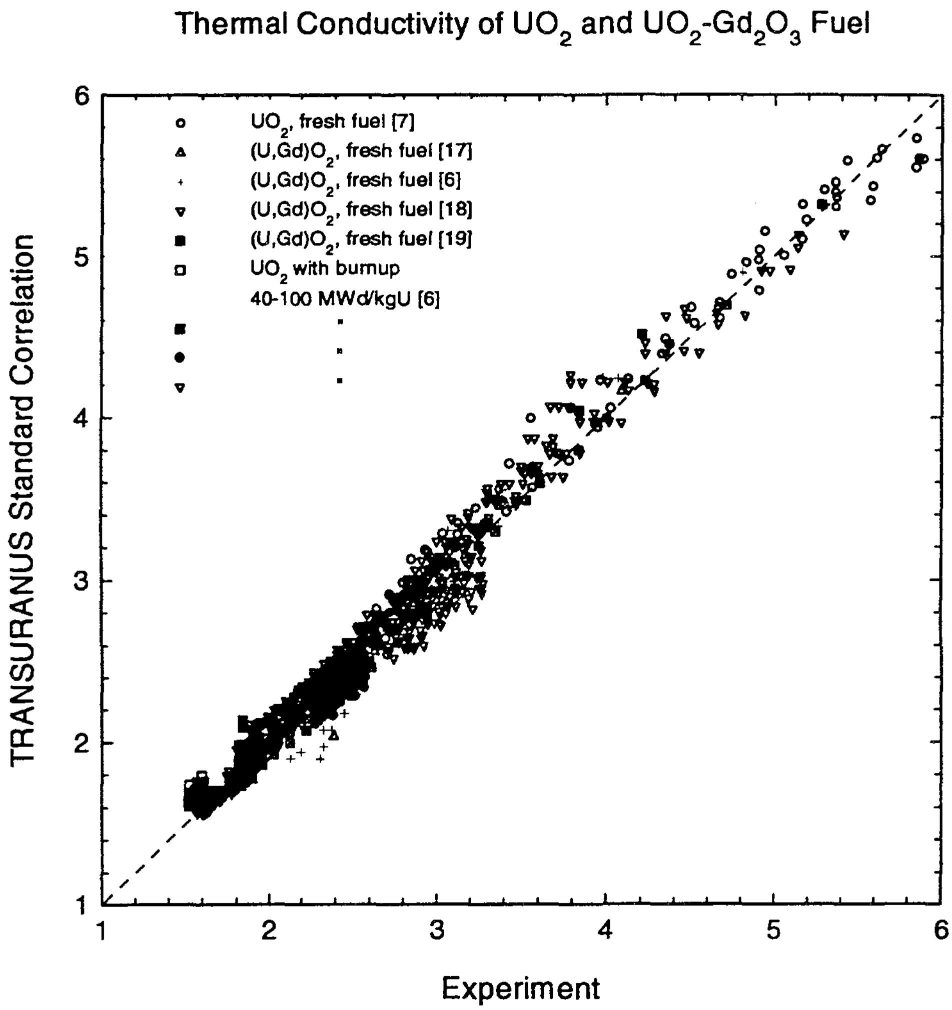


Figure 4. Comparison between measured and evaluated thermal conductivities.

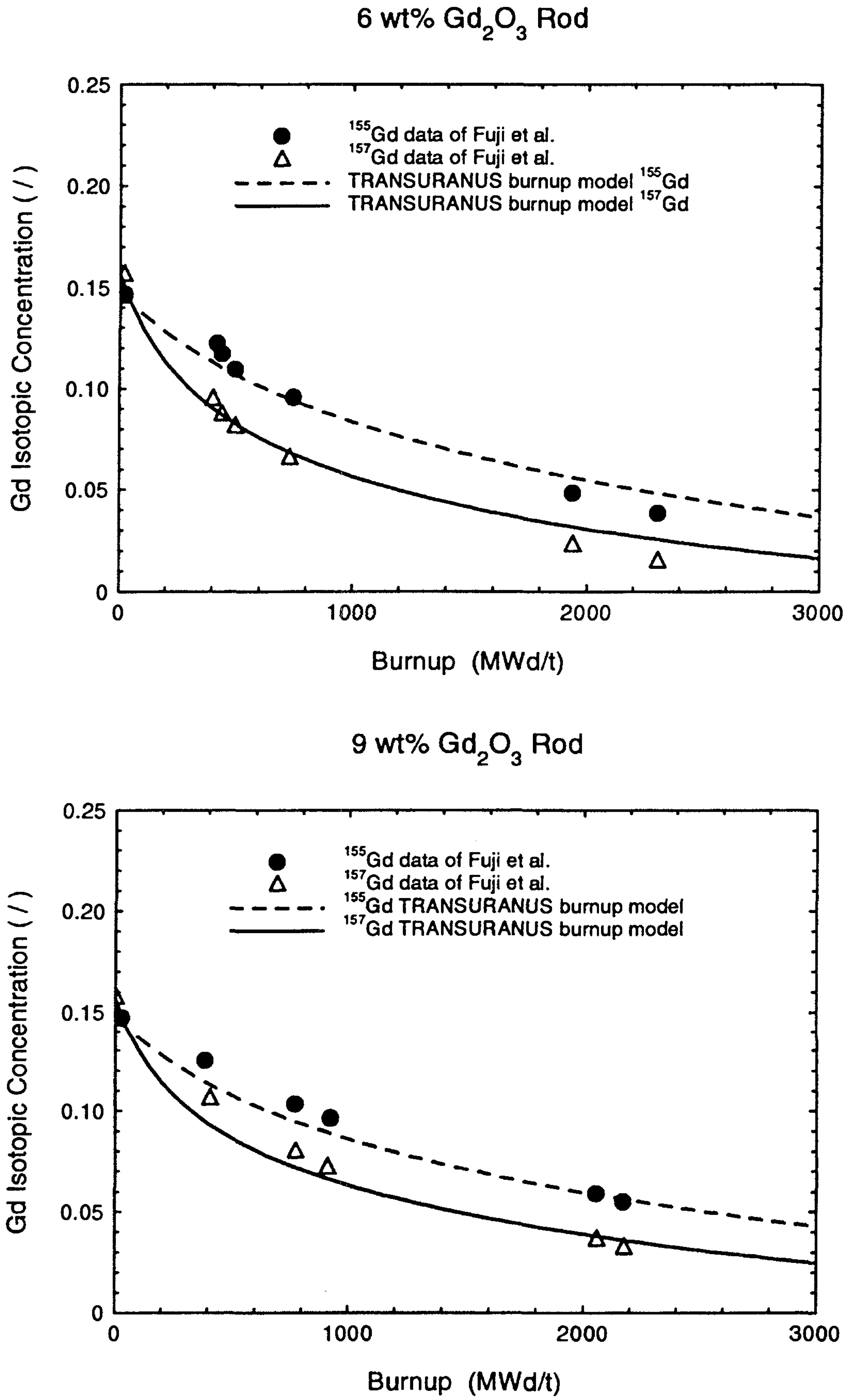


Figure 5. Average isotopic concentration of ^{155}Gd and ^{157}Gd in UO_2 fuels with 6 and 9 wt% Gd_2O_3 as a function of burnup. Comparison between the data of Fuji et al. [22] and the TRANSURANUS burnup model.

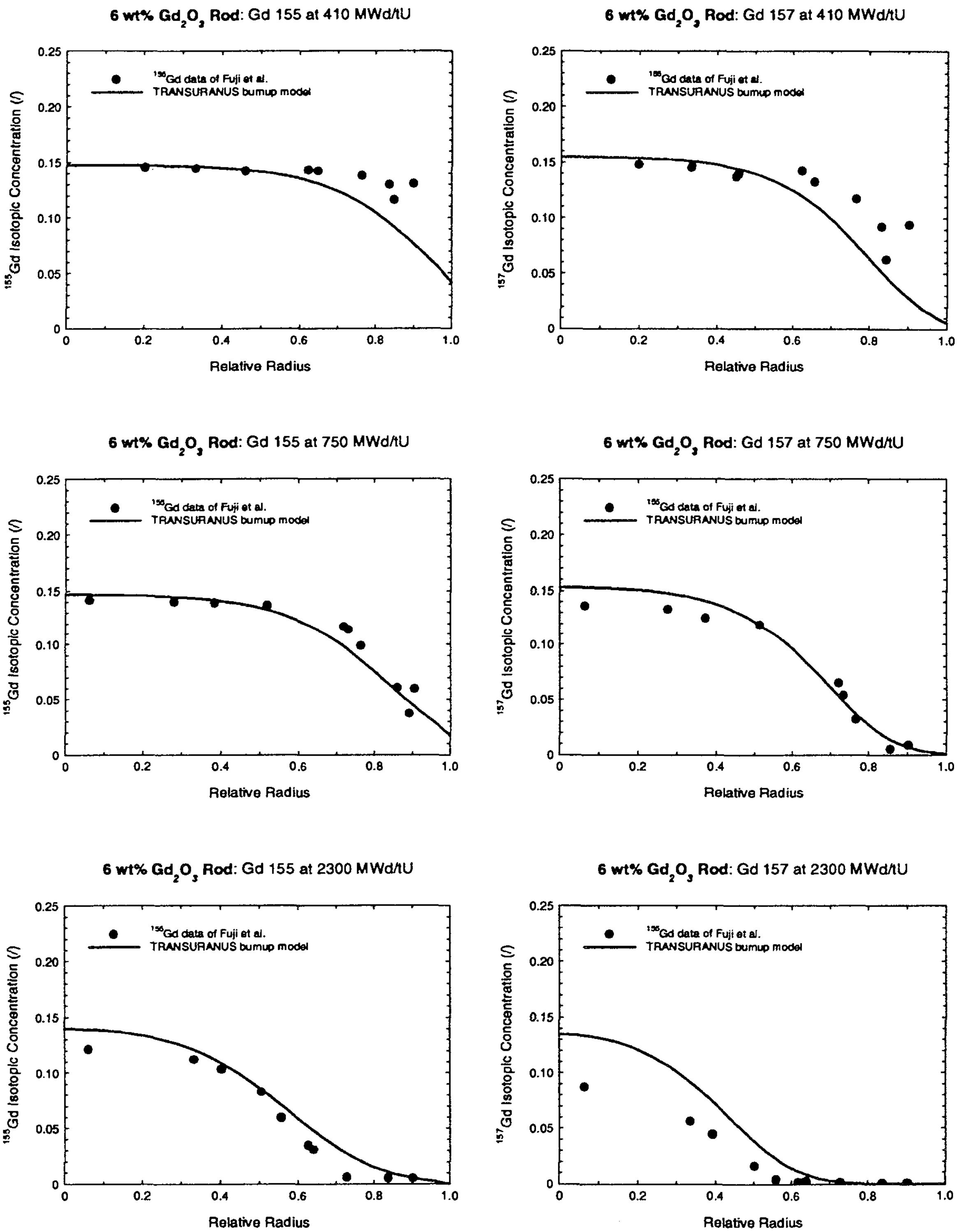


Figure 6. Radial distribution of ^{155}Gd and ^{157}Gd for different burnups; comparison between the data of Fuji et al. [22] and the TRANSURANUS burnup model. The initial concentration was 6 wt% of natural Gd.

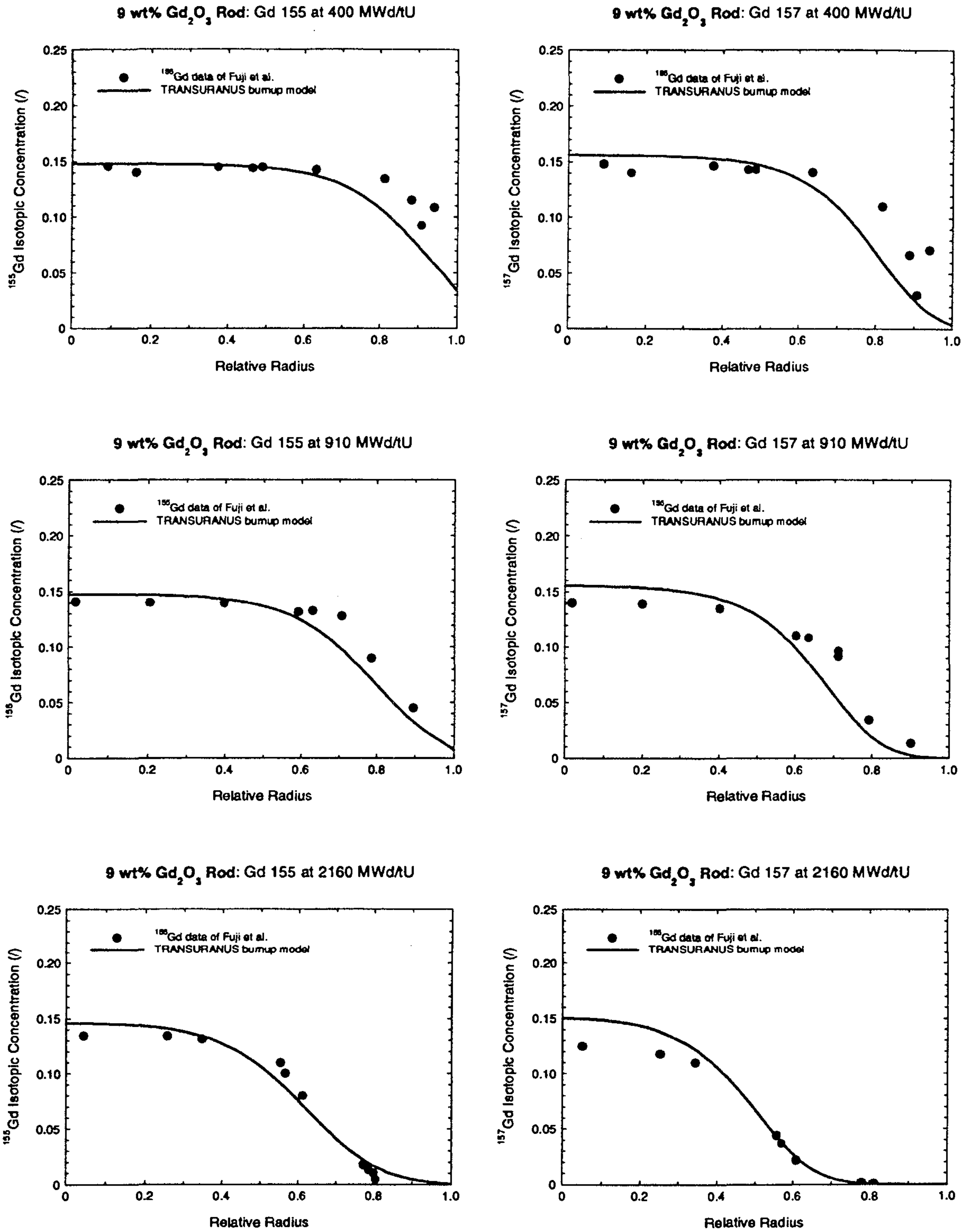


Figure 7: Radial distribution of ^{155}Gd and ^{157}Gd for different burnups; comparison between the data of Fuji et al. [22] and the TRANSURANUS burnup model. The initial concentration was 9 wt% of natural Gd.

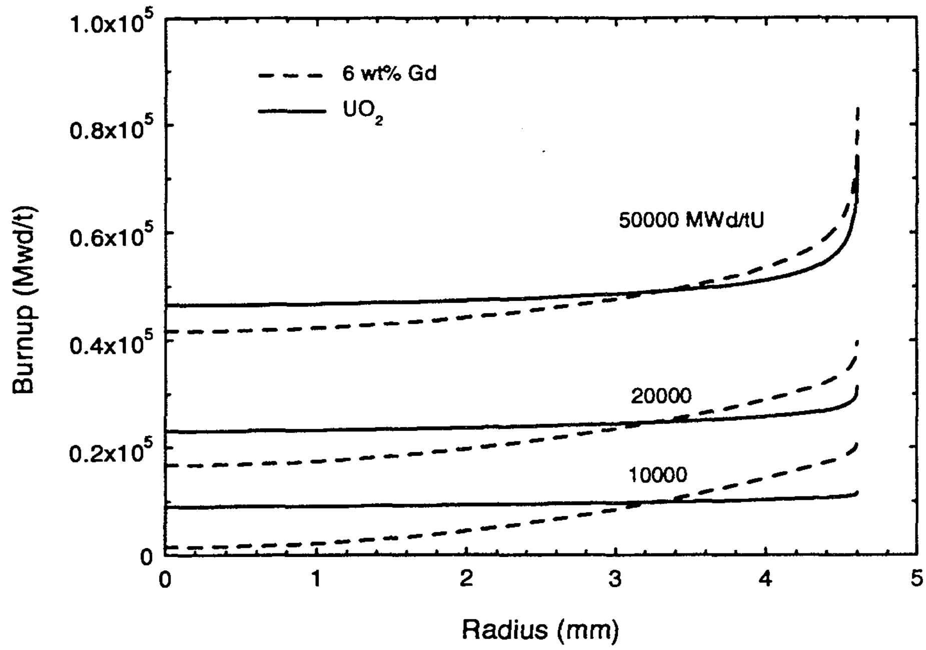


Figure 8. Comparison between the radial burnup profiles of a UO_2 and a $(U,Gd)O_2$ fuel (typical LWR fuel irradiated in the Halden reactor).

Differences between radial burnup profiles of standard UO_2 and Gd fuel are shown by way of example in Figure 8. As expected, the differences are pronounced at low burnup but even at a burnup of 50000 MWd/tU still visible.

3.2. Zirconium diboride (ZrB_2)

Developed by Westinghouse under the name IFBA (Integral Fuel Burnable Absorber), this fuel consists of a thin layer ZrB_2 ($\approx$ 5 µm) deposited by sputtering on the surface of the UO_2 pellets. The resulting ^{10}B loading is 1.7 mg ^{10}B per cm and the layer adheres perfectly to the UO_2 substrate [20, page 12]. This burnable absorber in the form of a thin layer works through the (n, α) reaction, e.g. He is produced. As a result there is a need to reduce the pre-pressurization level of the fuel rod.

The methodology to describe the neutron absorption of ^{10}B is the same as in the case of Gadolinia.

4. FISSION GAS RELEASE

It is commonly observed that fuel operated at "normal" rating shows very little fission gas release even up to high levels of burnup. Above 40 000 MWd/tU an enhancement takes place that could limit the lifetime of a fuel rod. In the following we take as an example Russian irradiation data of the KOLA-3 plant from the IFPE Database [23]. The data suggest that fission gas release below 40 000 MWd/tU is around 0.5% and rises linearly from 0.5% at 40000 MWd/tU to 1-2% at 50000 MWd/tU and $\approx$ 3% at 55000 MWd/tU.

Most theoretical fission gas release models are based on the assumptions that fission gas atoms diffuse inside the grain or precipitate into intra- and intergranular bubbles. Finally, they may reach the free pin volume basically by interlinkage of intergranular bubbles and subsequent venting of the grain boundary inventory. As a consequence, fission gas is released after an incubation time depending on the temperature. This is the physical understanding behind the so-called Vitanza threshold, which states that as long as the temperature remains below this threshold temperature, fission gas release does not occur or is insignificant.

The general problem is that these mechanisms do not always explain the enhancement of fission gas release above 40 000 MWd/tU. In most irradiations the rating and hence the temperature decrease with burnup. Diffusion processes may be insufficient to account for the enhancement of fission gas release and therefore the question is:

"Where does the enhanced fission gas release that is observed at extended burnup in all reactor irradiations come from"?

- From the inner hot regions by thermal processes or
- By an athermal process from the outer cold region that exhibits the high burnup structure?

The KOLA-3 data offers a unique opportunity to investigate this question further because

- The rating (and therefore the fuel temperature) is rather low and decreases with burnup.
- The use of annular UO_2 pellets further decreases the maximum fuel temperature compared with solid pellets.

The analyses confirmed that the maximum temperatures stay well below 1000°C. As expected, for all KOLA-3 rods analysed a small fission gas release of 0.3-0.4% at EOL is predicted [24]. The standard TRANSURANUS models cannot predict the enhancement of fission gas release above 40 000 MWd/tU for the KOLA-3 rods. In order to understand why this is the case, the calculated temperatures in different fuel sections have been compared with the Vitanza threshold. As can be seen in Figure 9 throughout the irradiation, the fuel temperatures remained well below the Vitanza threshold. This means, that either the calculated temperatures are too low, or the diffusion coefficient is too low, or a yet unknown fission gas release mechanism exists.

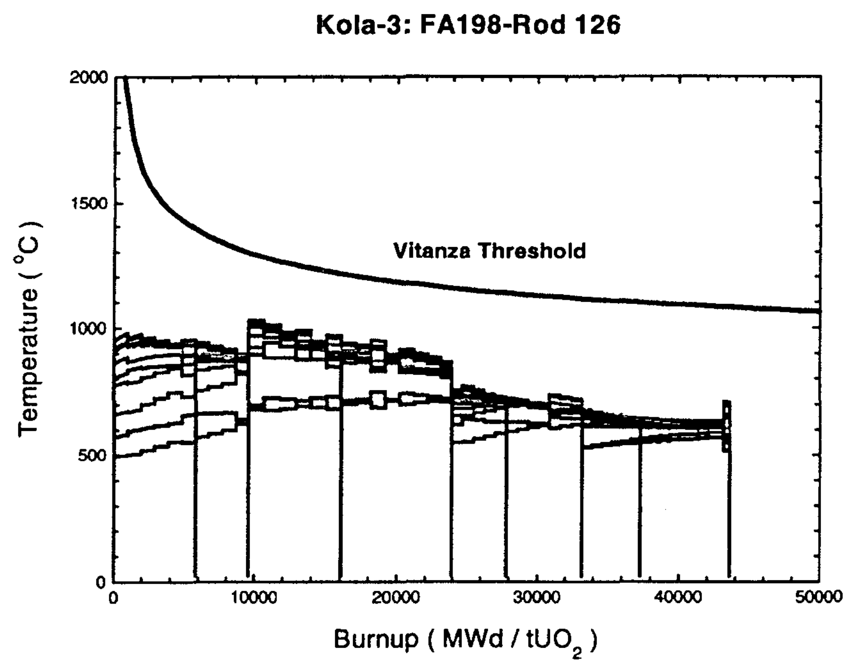


Figure 9. Comparison between the maximum fuel temperatures in different sections of rod 126 (fuel assembly 198) as calculated by the TRANSURANUS code with the Vitanza threshold. Note that the burnup is given in MWd/tUO₂.

This problem is not new and has also been found in Western fuel rods. We have analysed 2 hypotheses:

- During the formation of the High Burnup Structure, where a Xe-depletion of the matrix is observed, only part of the fission gas is released to pores inside the fuel, whereas the rest is released to the free volume. Glatz and Sätmark [25] have tried to answer this question by dissolving pellets of different shape in a sealed capsule. Fuel rod sections were cut from a UO_2 fuel rod irradiated in a power reactor to a burnup of 80000 MWd/tU. The fuel rod sections analysed were of different geometry: (a) a solid fuel pellet and b) two hollow sections with radii of $0.54 \le r/r_o \le 1$ and $0.91 \le r/r_o \le 1$, respectively. The conclusion is that in regions exhibiting the High Burnup Structure some of the fission gas is released. Since such experiments are extremely difficult, it is planned to repeat them with a slightly modified capsule to confirm the finding. A similar conclusion, however, is drawn by Walker from XRF analyses [26].
- From the many ITU EPMA data on high burnup fuels (for instance [27]) a constant Xe concentration of 0.2 to 0.3 wt% was found in the High Burnup Structure. This suggests that equilibrium exists between the created and the released Xe atoms given by

$$\bar{c}_i = \frac{b}{15 \frac{D_{Xe}}{a^2}} \approx 0.2 - 0.3 \quad wt\% \tag{4}$$

  where b is the creation rate, D_{Xe} the effective (apparent) diffusion coefficient and a is the grain radius [28]. Equation (4) allows to estimate D_{Xe} in the High Burnup Structure to be of the order of 10^{-22} m^2/s, which is an estimation for the athermal diffusion coefficient. Thus the second hypothesis for the enhanced fission gas release at high burnup is a simple irradiation enhanced (athermal) diffusion process. In order to investigate this effect, we have varied D_{Xe}.

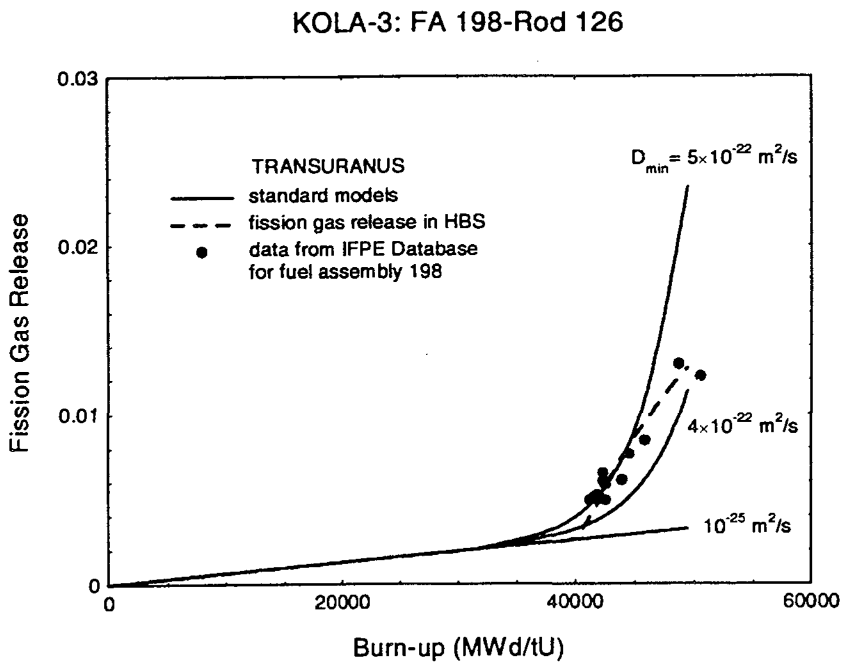


Figure 10. Fission gas release as a function of the rod average burnup for rod 126 of fuel assembly 198 (KOLA-3 irradiation) as calculated by the TRANSURANUS code by a) standard models with varying D_{Xe} and b) the assumption of a fission gas release from the High Burnup Structure.

Figure *10* shows that both hypotheses are in principle able to predict the measured fission gas release data in the KOLA-3 rods. The second hypothesis is also able to predict the Xe degradation in the High Burnup Stucture after grain subdivision (Figure 11). However, it is interesting to cross-check both hypotheses with measurements of the Xe to Kr ratio in the plenum. This ratio allows the origin of the fission gas released to be determined. Based on the assumption that the diffusion of Xe and Kr are identical, a relatively low ratio would indicate a release from the inner parts (more fissions of U than Pu) whereas a high ratio would indicate a release from the outer parts (more fissions of Pu than U). Unfortunately, no Xe to Kr data are available for the KOLA-3 data and we have compared the calculations with measurements performed by Toscano [31] at ITU on Western fuels. These experimental results indicate a rather low Xe to Kr ratio even at very high burnup which would indicate that the released fission gas does not originate from the outer parts. Figure 12 compares the data with the TRANSURANUS hypothesis that most of the released fission gas originates from the (outer) High Burnup Structure. For this mechanism the formation of a High Burnup Structure is essential, but once it has formed, the Xe to Kr ratio would increase significantly, clearly in contrast to the measured Xe to Kr ratios.

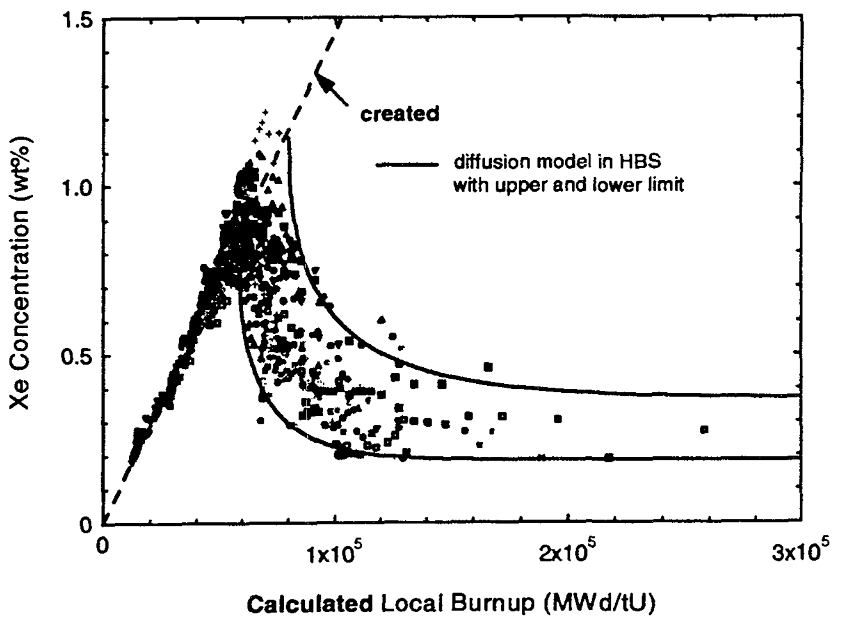


Figure 11. Comparison between measured EPMA data of C.T. Walker [29, 30] and predicted Xe concentraion in UO_2 fuel. The predictions are based on a diffusion process in the High Burnup Structure after grain subdivision.

Alternatively, the TRANSURANUS standard models are in good agreement with the measurements which would indicate that fission gas release originates more or less from the whole fuel (Figure 13).

The conclusion is evident: The standard TRANSURANUS fission gas release model can only explain the enhanced fission gas release above 40000 MWd/tU with a minimum (athermal) diffusion coefficient of the order of $10^{-22}\ m^2/s$. The assumption that most of the fission gas release stems from the High Burnup Structure must be questioned. Further research is needed to better understand the enhanced fission gas release at high burnup. This statement is valid for fuel rods of Western design as well as for WWER fuel rods.

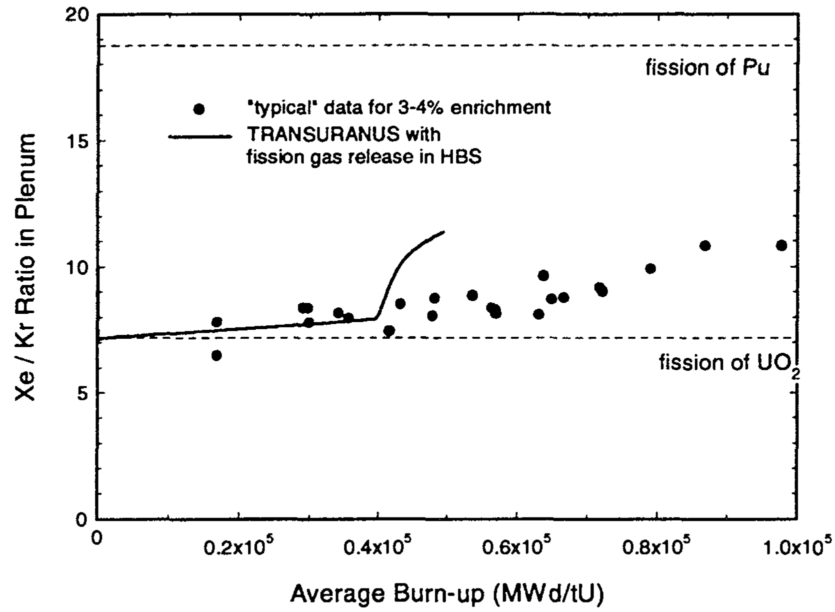


Figure 12. Xe/Kr ratio as a function of the rod average burnup for rod 126 of fuel assembly 198 (KOLA-3 irradiation) as calculated by the TRANSURANUS code on the assumption of a fission gas release from the High Burnup Structure. The data of Toscano [31] are included for the purpose of comparison.

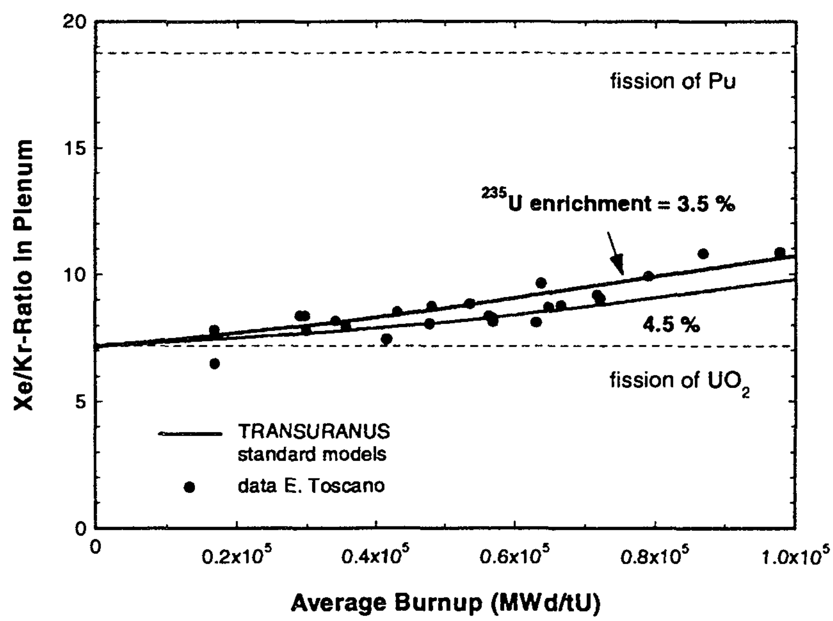


Figure 13. Xe/Kr ratio as a function of the rod average burnup for a "typical" rod as calculated by the TRANSURANUS code employing standard models. The predicted trends are in good agreement with the data of Toscano [31].

5. CONCLUSIONS

Two conclusions are drawn:

- The porosity in the High Burnup Structure needs clarification since the expansion of the fuel due to the formation of porosity as well as the decrease of the thermal conductivity are relevant for the thermal and mechanical behaviour.
- It seems that the standard TRANSURANUS fission gas release model can explain the enhanced fission gas release above 40 000 MWd/tU if a minimum (athermal) diffusion coefficient of the order of 10^{-22} m^2/s is applied. The assumption that most of the fission gas release stems from the High Burnup Structure must be questioned. Further research is needed to better understand the enhanced fission gas release at high burnup.

ACKNOWLEDGEMENTS

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Appendix

FURTHER TRANSURANUS DEVELOPMENTS

MONTE CARLO STATISTIC

The Monte Carlo statistics was extended to include several different probability density functions and several further stochastic variables. In addition to the normal (Gaussian) distribution it is now possible to apply log-normal, uniform (rectangle) as well as Cauchy distributions. Each individual distribution can be cutoff, i.e. a minimum and a maximum value can be considered. The new program "DISTRIB" in which the different distributions are programmed can easily be extended to include more types of distributions in the future.

INCORPORATION OF BROYDEN'S METHOD

General methods for the solution of the highly non-linear equations for creep and plasticity in fuel and cladding have been analysed [32]. The standard Newton's multidimensional method solves the set of equations

$$\begin{vmatrix} F_1(x_1, x_2, \ldots, x_n) \\ F_2(x_1, x_2, \ldots, x_n) \\ \ldots \\ \ldots \\ F_n(x_1, x_2, \ldots, x_n) \end{vmatrix} \quad \mathbf{F}(\mathbf{x}) \quad \mathbf{0}$$

by an iteration process

$$\mathbf{x}_{new} \quad \mathbf{x}_{old} \quad \mathbf{dx}$$

where

$$\mathbf{J}\,\mathbf{dx} \quad - \mathbf{F}$$

Here $\mathbf{J}$ is the Jacobian matrix.

Unfortunately, it is impossible to derive the Jacobian matrix for the mechanical equations used in the TRANSURANUS code [33] either analytically or approximately. Therefore, our research concentrated on the Broyden's method which is a multidimensional secant method. The Jacobian matrix $\mathbf{J}$ is approximated by the Broyden's matrix $\mathbf{B}$:

$$\mathbf{B}\,\mathbf{dx} \quad - \mathbf{F}$$

The interesting feature of Broyden's method is that the Broyden's matrix is updated during the iteration process:

$$\mathbf{B}_{i\,1} \quad \mathbf{B}_i \quad \frac{(\mathbf{dF}_i - \mathbf{B}_i\,\mathbf{dx}_i) \otimes \mathbf{dx}_i}{\mathbf{dx}_i\,\mathbf{dx}_i}$$

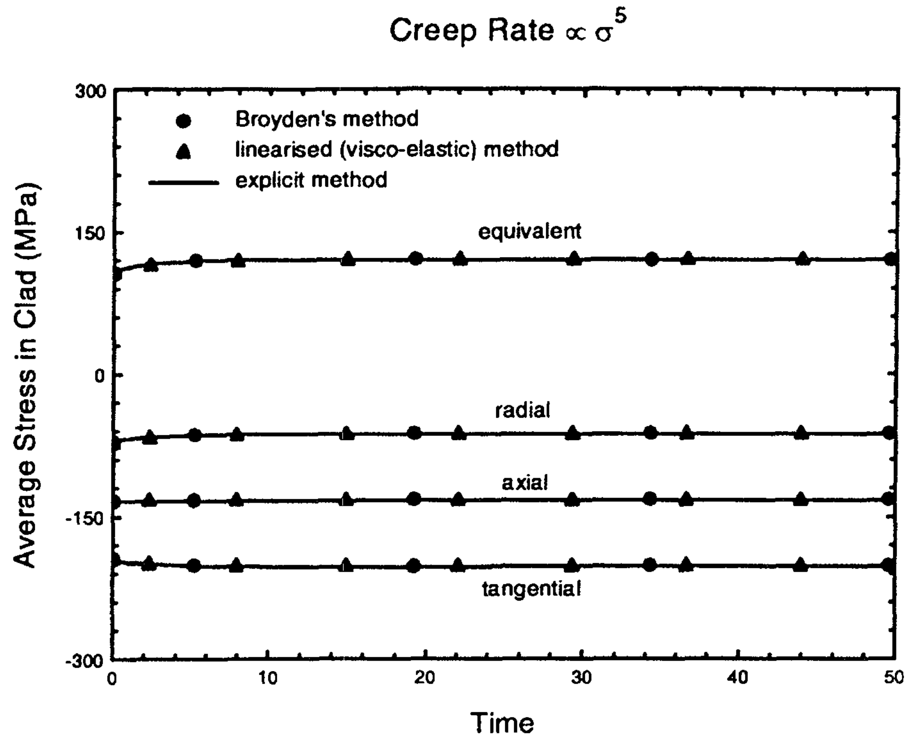

Figure 14. Average stresses in a cladding under outer pressure as a function of time for a highly non-linear creep law (creep rate ∝ s^{5}_{eff}). Shown is the comparison between different methods.

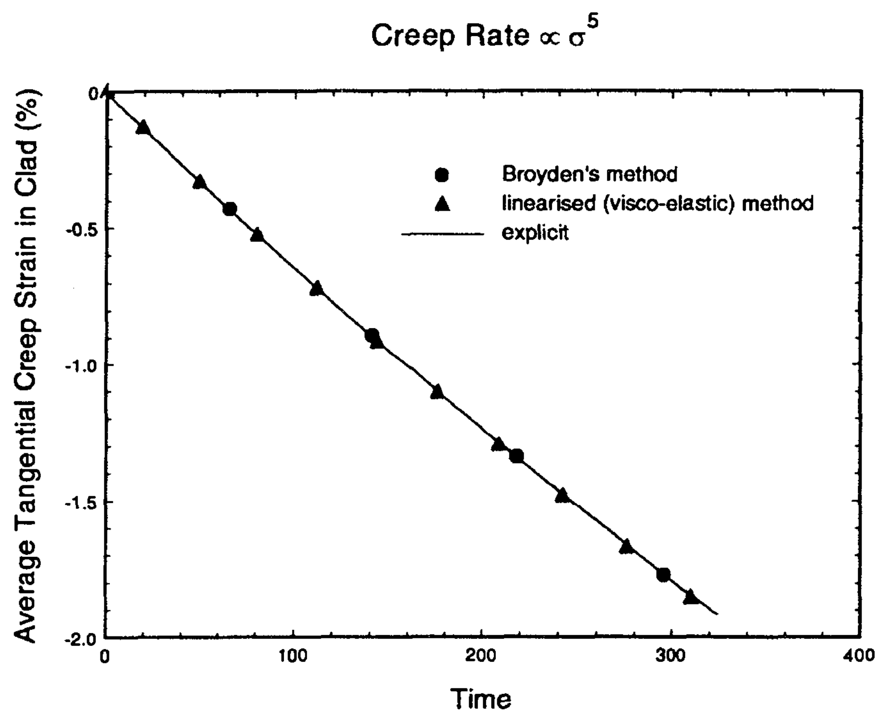

Figure 15. Average tangential creep strain in a cladding under outer pressure as a function of time for a highly non-linear creep law (creep rate ∝ s^{5}_{eff}). Shown is the comparison between different methods.

where $d\mathbf{F}_i \; \mathbf{F}_{i+1} - \mathbf{F}_i$, $d\mathbf{x} \; \mathbf{x}_{i+1} - \mathbf{x}_i$ and the symbols "·" and "×" denote the scalar and vector product, respectively. Various variants of Broyden's method have been studied and the most promising has been incorporated into the TRANSURANUS code. First tests confirmed excellent agreement with the existing explicit and linearised (visco-elastic) treatments. Two examples are given in Figure 14 and Figure 15. Note that in Figure 14 the average stresses need to be approximately constant due to the equilibrium condition. This is not the case for local stresses not shown here. Further tests will be necessary to study time step control (convergence) and efficiency.

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