Journal of Nuclear Materials 208 (1994) 223–231
North-Holland

journal of nuclear materials

The radial distribution of plutonium in high burnup UO_2 fuels

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Received 9 August 1993; accepted 12 October 1993

A new model (TUBRNP) is described which predicts the radial power density distribution as a function of burnup (and hence the radial burnup profile as a function of time) together with the radial profile of uranium and plutonium isotopes. Comparisons between measurements and the predictions of the TUBRNP model are made on fuels with enrichments in the range 2.9 to 8.25% and with burnups between 21000 and 64000 MWd/t. It is shown to be in excellent agreement with experimental measurements and is a marked improvement on earlier versions.

1. Introduction

The prediction of fuel rod behaviour in a Light Water Reactor (LWR) at high burnup is difficult because the thermal and mechanical analysis depend strongly on complex material behaviour that varies with burnup. In particular the radial power density distribution in fuel rods is nonuniform and is a function of burnup. This paper investigates the dependence of the radial (local) power density q''' on the burnup which is basically determined by the fuel rod geometry, the initial concentration of fissile material and the Pu buildup. At the beginning of irradiation, i.e. at low burnup, the concentration of fissile material is constant which means that the radial power has a relatively small variation across the radius. At high burnup there is a variation in this concentration with a marked increase in the ^{239}Pu concentration near the fuel surface. This increases with burnup and must be taken into account in the thermal analysis [1]. The variation can be explained as follows: due to the capture of epithermal neutrons in the resonances of ^{238}U (with a very high absorption cross section) the Pu buildup near the pellet surface is larger than at the centre of the fuel. Consequently, the power density profile which is proportional to the neutron flux and the concentration of fissile material is steeper near the pellet surface at high burnup. This problem has been studied before by other workers in this field, Palmer et al., Carlsen and Sah, Kleykamp, Matsumura and Kameyama [2–5].

The model presented here can be considered as an extension of the well known RADAR (RAting Depression Analysis Routine) model of Palmer et al. [2]. This new model predicts the radial power density distribution as a function of burnup (and hence the radial burnup profile as a function of time) together with the radial profiles of ^{235}U, ^{238}U and the Pu isotopes ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu. The model is included in the TRANSURANUS code [6] and is named TUBRNP (TRANSURANUS burnup model). This work describes the basic assumptions made, the database used to develop the model parameters and comparison with experiment.

2. Theory

The original RADAR model of Palmer et al. [2] consists of: (1) a differential equation for the local ^{235}U concentration, (2) a differential equation for the local ^{239}Pu concentration, (3) the solution of simple diffusion theory for the thermal neutron flux. In the RADAR model the plutonium is distributed radially according to an empirically determined radial shape function. The main input parameters of the RADAR model are the geometry of the fuel, the initial concentration of ^{235}U, the leakage factor L and the resonance escape probability p.

In recent years the RADAR model was applied extensively within the TRANSURANUS code and gave

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good results for low and medium burnup. At high burnup (> 4 at%) the results were not so good for the following two reasons: (a) for PWR conditions with a recommended resonance escape probability of $p \approx 0.65$ the ^{239}Pu concentration is overestimated, (b) the buildup of higher Pu isotopes is not taken into account. At high burnup (> 4 at%) the ^{239}Pu concentration is less than 60% of the total Pu concentration.

In order to overcome these limitations the basic equations of the RADAR model were extended to include also the higher Pu isotopes. Following the approach of the codes ORIGEN by Ball [7] and KORIGEN by Fischer and Wiese [8] the equations which give the average concentrations in the fuel are

$$\frac{\mathrm{d}\bar{N}_{235}}{\mathrm{d}t} = -\sigma_{\mathrm{a},235}\bar{N}_{235}\phi, \tag{1a}$$

$$\frac{\mathrm{d}\bar{N}_{238}}{\mathrm{d}t} = -\sigma_{\mathrm{a},238}\bar{N}_{238}\phi, \tag{1b}$$

$$\frac{\mathrm{d}\bar{N}_{j}}{\mathrm{d}t} = -\sigma_{\mathrm{a},j}\bar{N}_{j}\phi + \sigma_{\mathrm{c},j-1}\bar{N}_{j-1}\phi, \tag{1c}$$

where $\bar{N}$ is the number of atoms per unit volume, σ_{a} the absorption cross section, σ_{c} the capture cross section, ϕ the neutron flux, and the subscript j represents the isotopes ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu.

In a fuel rod performance code such as TRANSURANUS the linear rating is usually prescribed and from this the burnup is determined. Thus it is favourable to convert the increment of the fluence $\phi\Delta t$ to a burnup increment Δbu:

$$\Delta\mathrm{bu} = \frac{q'''\Delta t}{\rho_{\mathrm{fuel}}} = \frac{\alpha}{\rho_{\mathrm{fuel}}}\sum_k \sigma_{\mathrm{f},k}\bar{N}_k\phi\Delta t, \tag{2}$$

where q''', is the power density, ρ_{fuel} the fuel density, $\sigma_{\mathrm{f},k}$ the fission cross section of isotope k and α is a conversion factor. The quantity $\Sigma_k\sigma_{\mathrm{f},k}\bar{N}_k\phi$ is the number of fissions per unit volume per unit time. Each fission event generates approximately 200 MeV. It is assumed that the fuel is UO_2 and that the burnup is given in MWd per tonne of heavy metal (MWd/t). The conversion factor α then becomes $\alpha = 3.35 \times 10^{-16}$ and the basic equations can be written as,

$$\frac{\mathrm{d}\bar{N}_{235}}{\mathrm{dbu}} = -\sigma_{\mathrm{a},235}\bar{N}_{235}A, \tag{3a}$$

$$\frac{\mathrm{d}\bar{N}_{238}}{\mathrm{dbu}} = -\sigma_{\mathrm{a},238}\bar{N}_{238}A, \tag{3b}$$

$$\frac{\mathrm{d}\bar{N}_{j}}{\mathrm{dbu}} = -\sigma_{\mathrm{a},j}\bar{N}_{j}A + \sigma_{\mathrm{c},j-1}\bar{N}_{j-1}A, \tag{3c}$$

where

$$A = 0.8815\frac{\rho_{\mathrm{fuel}}}{\alpha\sum_k \sigma_{\mathrm{f},k}\bar{N}_k}.$$

Note that the cross sections in the equations above are to be considered average effective cross sections valid for a specific reactor type: here we are interested in PWR and BWR conditions. In view of the simplifications made so far (one neutron group, spectrum averaged cross sections and "selected isotopes") a common set of cross sections for both types of reactor is justified. Analyses performed with the KORIGEN code support the assertion that the differences between PWR and BWR conditions are not significant.

3. Local concentrations and radial profiles

So far, the basic equations were formulated as an average balance in the fuel. To derive local concentrations consider the following set of equations:

$$\frac{\mathrm{d}N_{235}(r)}{\mathrm{dbu}} = -\sigma_{\mathrm{a},235}N_{235}(r)A, \tag{4a}$$

$$\frac{\mathrm{d}N_{238}(r)}{\mathrm{dbu}} = -\sigma_{\mathrm{a},238}\bar{N}_{238}f(r)A, \tag{4b}$$

$$\frac{\mathrm{d}N_{239}(r)}{\mathrm{dbu}} = -\sigma_{\mathrm{a},239}N_{239}(r)A + \sigma_{\mathrm{c},238}\bar{N}_{238}f(r)A, \tag{4c}$$

$$\frac{\mathrm{d}N_{j}(r)}{\mathrm{dbu}} = -\sigma_{\mathrm{a},j}N_{j}(r)A + \sigma_{\mathrm{c},j-1}N_{j-1}(r)A. \tag{4d}$$

The local concentration of ^{238}U, $N_{238}(r)$, is written as $\bar{N}_{238}f(r)$, where $f(r)$ is a radial shape function with a normalisation factor defined by

$$2\frac{\int_{r_{\mathrm{in}}}^{r_{\mathrm{out}}} f(r)r\,\mathrm{d}r}{r_{\mathrm{out}}^2 - r_{\mathrm{in}}^2} = 1, \tag{5}$$

where r_{in} and r_{out} are the inner and outer fuel radii. The shape function takes into account the resonance absorption in ^{238}U that leads to the formation of ^{239}Pu. This distribution function can be interpreted as the combination of a constant production of ^{239}Pu from thermal neutron capture plus a highly nonlinear term for the production due to resonance absorption.

The local power density $q'''(r)$ which is needed for the thermal analysis is proportional to the neutron flux and the macroscopic cross section for fission,

$$q'''(r) \propto \sum_k \sigma_{\mathrm{f},k}N_k\phi. \tag{6}$$

In order to determine ϕ, thermal flux diffusion theory can be applied and the resulting solutions of the differential equation are

$$\phi(r) \propto I_0(\kappa r) \quad \text{for the solid cylinder,} \tag{7a}$$

$$\phi(r) \propto I_0(\kappa r) + \frac{I_1(\kappa r_{\mathrm{f,out}})}{K_1(\kappa r_{\mathrm{f,out}})} K_0(\kappa r)$$

$$\text{for the hollow cylinder,} \tag{7b}$$

where I and K are the modified Bessel functions. The inverse diffusion length, κ, is given by

$$\kappa = \sqrt{\Sigma_{\mathrm{a,tot}}/D}\,, \tag{8}$$

where

$$\Sigma_{\mathrm{a,tot}} \approx \sum_k \sigma_{\mathrm{a},k} \bar{N}_k\,,\ D = \frac{1}{3\Sigma_{\mathrm{s}}} = \frac{1}{3\sigma_{\mathrm{s}} \bar{N}_{\mathrm{tot}}}\,;$$

$\bar{N}_k$ and $\bar{N}_{\mathrm{tot}}$ are the averaged values.

For the nonlinear part of the distribution function, due to resonance absorption, the empirical radial shape function of Palmer et al. [2], $f(r) = 1 + 3\exp(-9.7\sqrt{r_{\mathrm{out}} - r})$, gave satisfactory results. A new form of this function has been developed which results in excellent agreement with experiment. This distribution function is of the form

$$f(r) = 1 + p_1 \exp\left(-p_2 (r_{\mathrm{out}} - r)^{p_3}\right), \tag{9}$$

where p_1, p_2 and p_3 are constants. The values of these parameters were derived after careful and exhaustive comparisons with measurements. The above set of equations have been programmed in a subroutine of TRANSURANUS called TUBRNP (TRANSURANUS burnup equations).

4. Experimental data and model parameters

Measurements of plutonium profiles have been done for many years [9] and is a topic of current interest [10]. The data used here comes from a variety of sources which acts as a good test for the general application of the model. The set of electron probe microanalysis data described below acts as a database that can be used to test the validity of the TUBRNP model. A small sample of the data was used to fit the model parameters. Comparisons were made with simulations using the ORIGEN and KORIGEN codes; also with those of a model developed by Matsumura and Kameyama [5]. These comparisons are discussed in section 5.

4.1. Fuel type and irradiation history

Fuel provided by the Nuclear Power Division of the Electricity Power Research Institute (EPRI) and General Electric (GE) was used to provide Pu data. The EPRI fuel specimens were of PWR design and had been irradiated in the BR3 reactor in Belgium to a burnup of 39400 to 63000 MWd/t. This fuel had seen a linear heat generation rate (LHGR) as high as 40.3 kW m^{-1}. The GE fuel specimens were of BWR design and had been irradiated in the Millstone-1 (USA) to a burnup between 23000 and 39000 MWd/tM. The irradiation of this fuel had spanned eight power cycles and lasted 2880 days. During this time the peak pellet LHGR fell from about 15 kW m^{-1} at the end of the third cycle to 8–10 kW m^{-1} in the eighth cycle.

Relevant pellet and pin design characteristics of the EPRI and GE fuel specimens referred to in this paper are shown in table 1. The principal difference between

Table 1
Pellet and in design characteristics of the EPRI and GE fuels

Design parameter	EPRI fuel				GE fuel STRO
	A1	A2	A3	A4	
Pellet diameter (mm)	8.04	8.04	8.04	8.04	10.9
Pellet density (%TD)	94.4	94.5	95.5	95.5	95.7
2D Grain size (mm)	11	11	10	10	12
Enrichment (% ^{235}U)	8.25	8.25	5.75	5.75	2.9
Fill gas [a]	He (2.0)	He (2.0)	He (2.0)	He (2.0)	He (1.7)
Diametrical gap (mm)	0.20	0.20	0.20	0.20	0.23
Cladding material	Zr-4	Zr-4	Zr-4	Zr-4	Zr-2 [b]

[a] The value in parenthesis is the pressure in MPa.
[b] With a Zr liner.

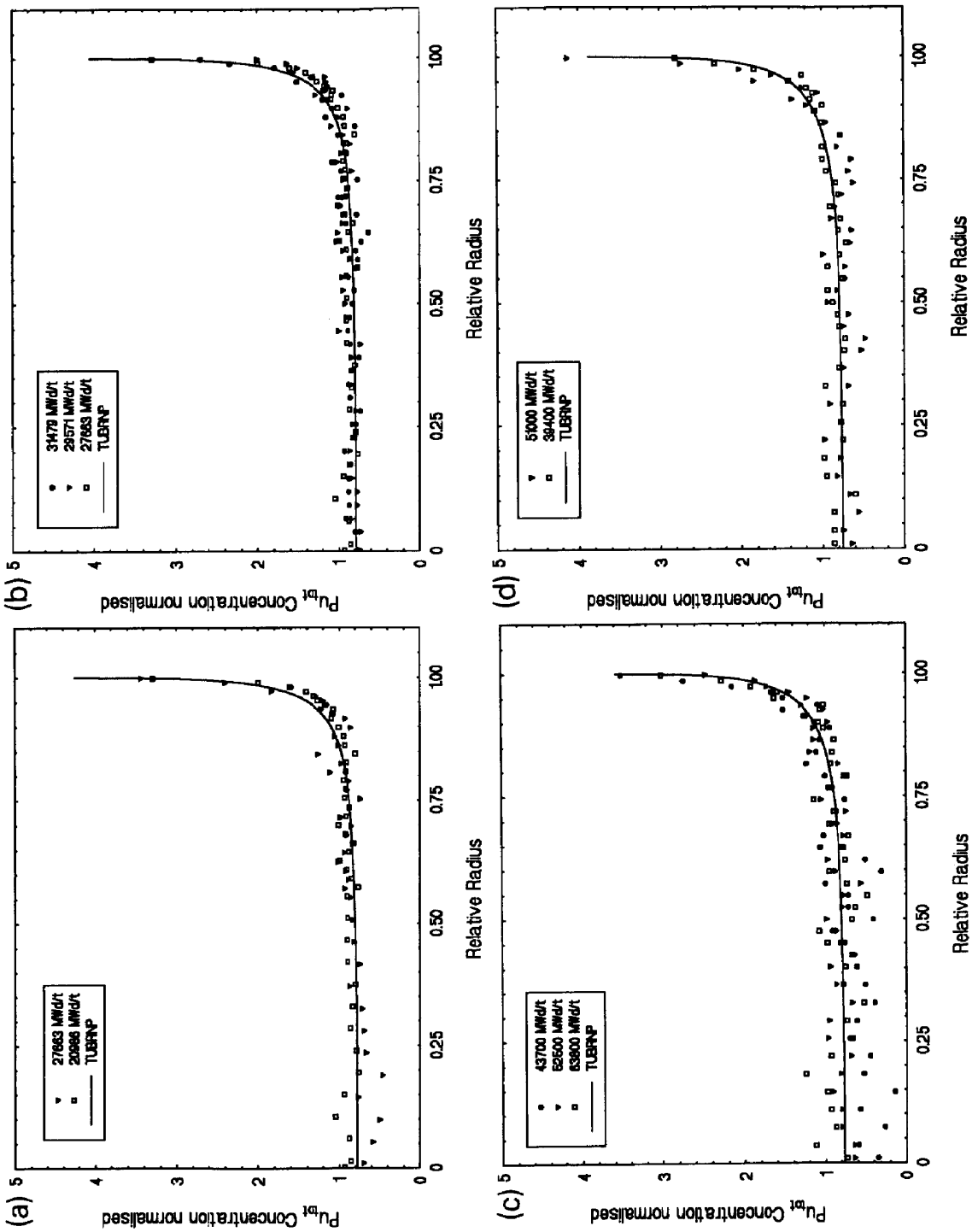

Fig. 1. (a) Comparison of the radial total Pu concentration measured in different STRO fuels (enrichment $^{235}U = 2.9\%$) with that calculated by the TUBRNP model for 25000 MWd/t. (b) Comparison of the radial total Pu concentration measured in different STRO fuels (enrichment $^{235}U = 2.9\%$) with that calculated by the TUBRNP model for 29000 MWd/t. (c) Comparison of the radial total Pu concentration measured in different EPRI fuels (enrichment $^{235}U = 5.75\%$) with that calculated by the TUBRNP model for 55000 MWd/t. (d) Comparison of the radial total Pu concentration measured in different EPRI fuels (enrichment $^{235}U = 8.25\%$) with that calculated by the TUBRNP model for 45000 MWd/t.

the two fuel types is the enrichment which was much lower in the case of the BWR fuel specimens.

4.2. Measurement of plutonium by electron probe microanalysis

Quantitative EPMA of Pu was carried out at an electron acceleration potential of 25 keV and a beam current of 100 nA using a PuO_2 standard. The need to reduce surface effects and to ensure good X-ray counting statistics dictates the use of a high acceleration potential. Applying the equation of Reed [11] it can be calculated that the depth of electron penetration in UO_2 at 25 keV is about 0.5 μm and that the diameter of X-ray excitation is about 3.5 μm. To decrease the radiation background the pulse height analyser was used in the differential mode. The conventional microprobe correction procedure was carried out using the Institute's QUAD2 program of Farthing et al. [12] which is based on the Quadrilaterial Model of Scott and Love [13].

Plutonium was analysed using the M_β X-ray line and a quartz 1011 diffracting crystal. The intensity of the Pu M_β line must be corrected for overlap with the neighbouring U M_{γ_2} line. To make this correction, the spectrometer was positioned on the Pu M_β line and the X-ray intensity obtained from the UO_2 standard at this setting was recorded. About 8 cps were registered which corresponds to 0.76 wt% Pu. At each spot analysed the true percentage of Pu was obtained by subtracting from the measured concentration an amount given by the product of the reference concentration of 0.76 wt% and the fraction of UO_2 present.

Depending on the fuel pellet diameter, 35 to 45 data points were used to construct the Pu concentration profile. These were measured at intervals of 50 to 250 μm along the pellet radius. At each location, the concentration of Pu was determined from the average of four peak and three background measurements of 25 s duration. The specimen current image (absorbed electron current) was used to obtain information about the fuel microstructure at the locations selected for analysis and to position the electron beam.

The confidence interval on a measured Pu concentration of 2.0 wt% is around 10% relative at a significance level of 99%. This uncertainty is due solely to the statistics of X-ray counting. After correction for the use of a compound standard and after accounting for X-ray contributions from the U M_{γ_2} line the confidence interval is of the order of 20% relative.

4.3. Fitting the model parameters

The cross sections used in the TUBRNP model were taken in a slightly modified form from KORIGEN and ORIGEN. The remaining parameters of

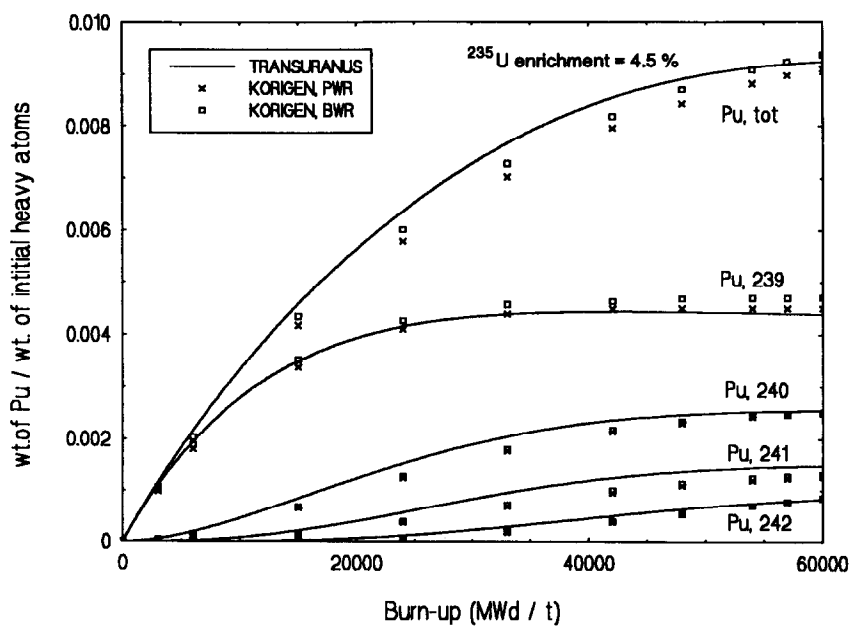

Fig. 2. The total average Pu concentration and constituent isotope concentrations in a PWR and BWR fuel calculated with the TUBRNP model (^{235}U enrichment is 4.5%). The predictions of the KORIGEN code are included for the purpose of comparison.

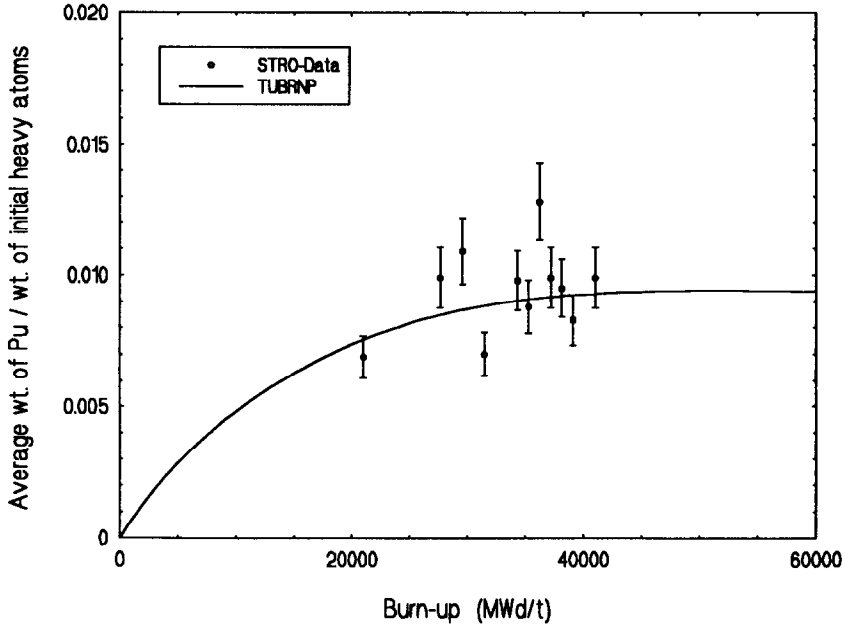

Fig. 3. Comparison of the average Pu concentration measured in different STRO fuels (enrichment $^{235}U = 2.9\%$) at different burnups with that calculated by the TUBRNP model.

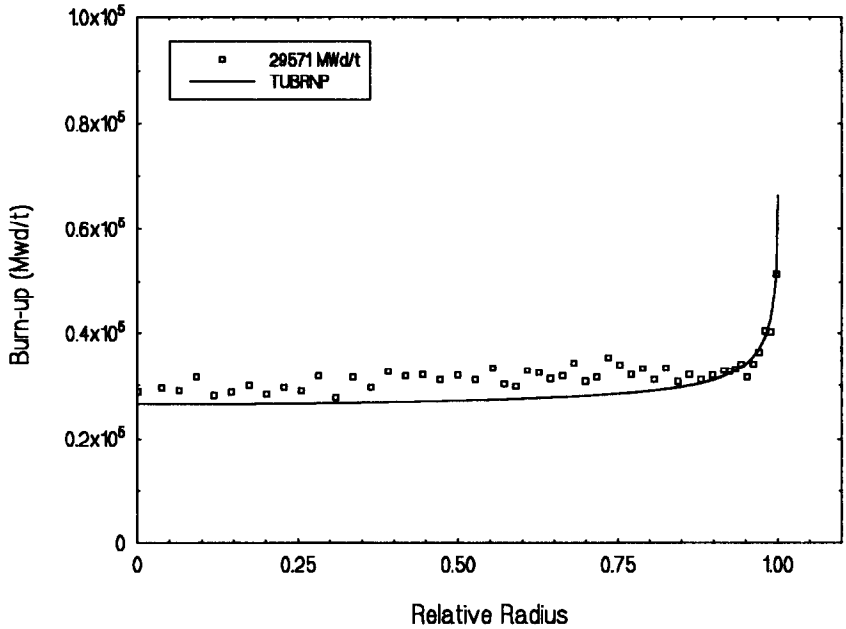

Fig. 4. Comparison of the measured radial burnup profile in a STRO fuel (enrichment $^{235}U = 2.9\%$; average burnup = 29571 MWd/t) with that calculated by the TUBRNP model.

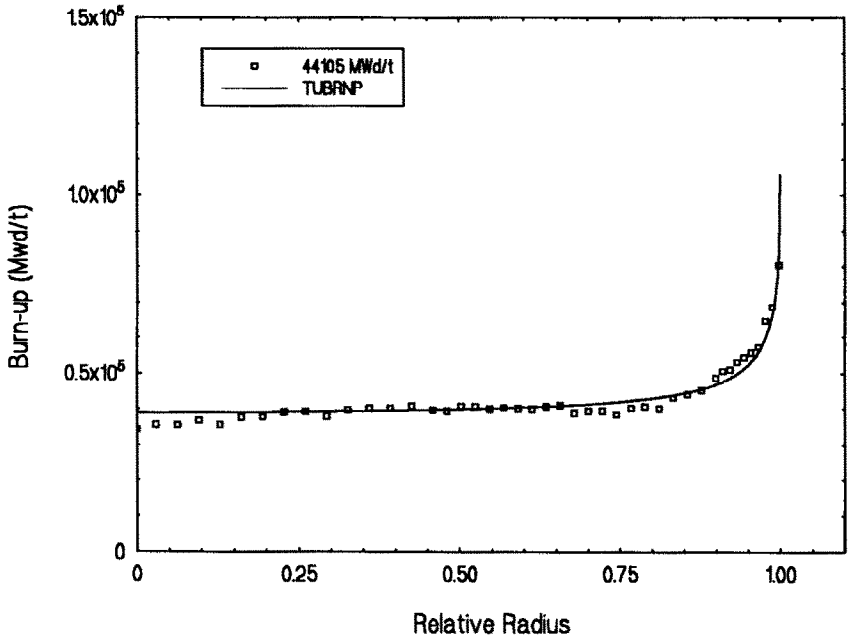

Fig. 5. Comparison of the measured radial burnup profile in a Risø fuel (enrichment $^{235}U = 2.95\%$; average burnup 44 105 MWd/t) with that calculated by the TUBRNP model.

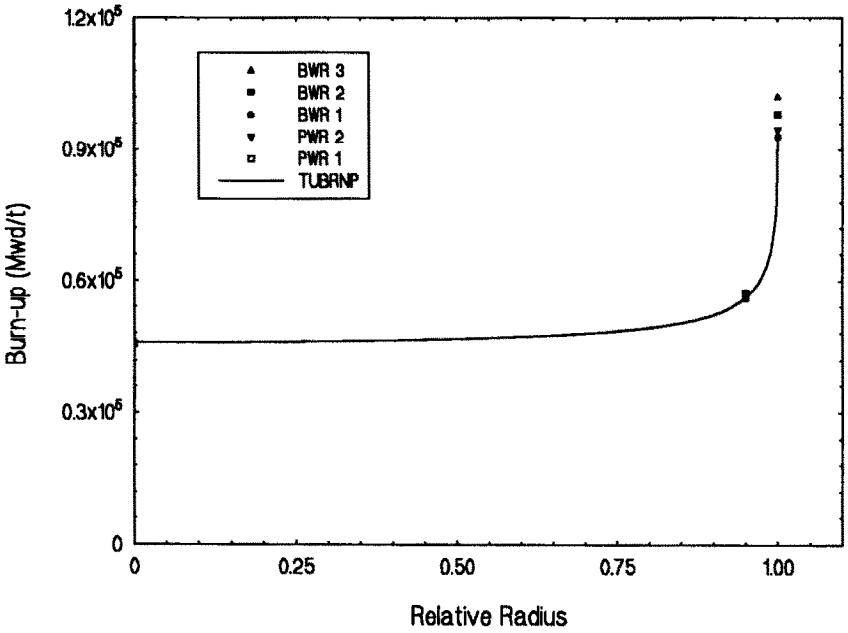

Fig. 6. Comparison of the radial burnup profile in several fuel types (enrichment $^{235}U = 5\%$) at 50000 MWd/t calculated by the model of Matsumura and Kameyama [5] with that of the TUBRNP model.

TUBRNP which determine the shape function (p_1, p_2 and p_3), eq. (9), were fitted to the data described above (measured radial Pu profiles from EPRI and GE fuels). These three fitted parameters were fixed in the model and the *same* ones were used for all other comparisons with experiment and other models. Given that these are the only "free" parameters, TUBRNP is a purely predictive model.

5. Prediction of measured data and comparison with other models

In figs. 1a–1d the radial dependence of the Pu predicted by the model is compared with the measurements. As mentioned above these comparisons were done using fuels with enrichments between 2.9 and 8.25% and with burnups between 21 000 and 64 000 MWd/t. It is important to note that in all cases the same parameters were used for the radial shape function, eq. (9). The agreement between the theoretical curve and the measured data is very good in all cases and in some is quite remarkable.

Although TUBRNP is a local model it must of course be able to describe the average concentrations of the different nuclides. The total Pu buildup in the fuel is strongly dependent on the ^{235}U enrichment. In contrast the enrichment is not a significant factor in determining the Pu profile at high burnup since (except for very large values) it will not affect the rate of epithermal neutron capture. An example of the comparison of the model curve with the predictions of the KORIGEN code is given in fig. 2.

Further verification of the TUBRNP model is illustrated in fig. 3. Here, the measured average Pu concentration at different burnups is compared with the model predictions. It should be noted that the average Pu concentration was derived from the interpretation of the local measurements. Given the difficult absolute measurement of local Pu concentrations and the simplicity of the model these results show no major discrepancies between theory and experiment.

Two more comparisons have been made as independent checks of the TUBRNP model. First, radial burnup profiles have been compared. As seen from figs. 4 and 5 there is close agreement between measurement and prediction. Secondly, the model was compared with the one developed by Matsumura and Kameyama

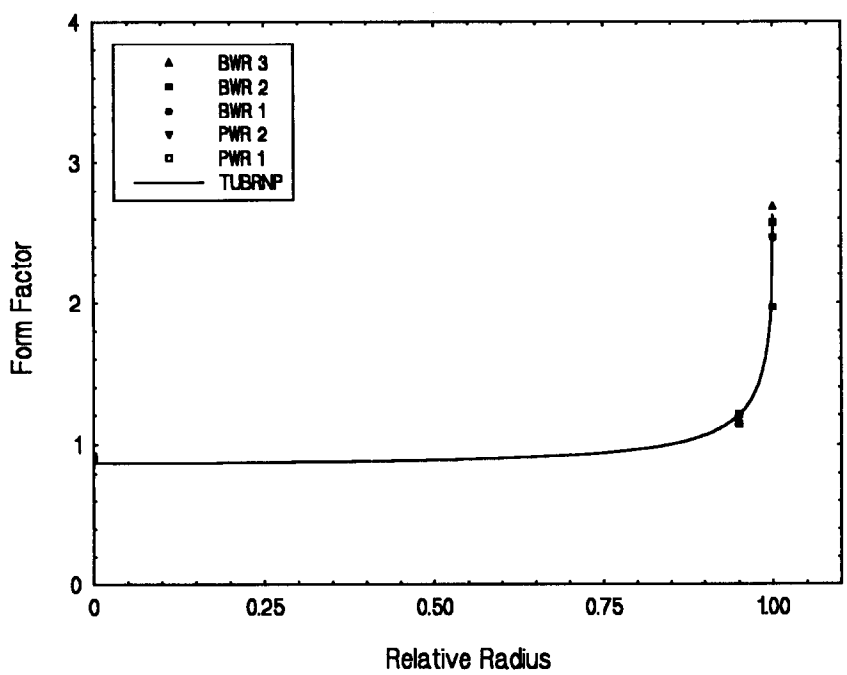

Fig. 7. Comparison of the radial power density distribution in several fuel types (enrichment $^{235}U = 5\%$) at 50 000 MWd/t calculated by the model of Matsumura and Kameyama [5] with that of the TUBRNP model.

[5]. Their model for the burnup and radial profile was fitted to experimental data from several fuels. The data came from both BWR and PWR fuel rods with varying enrichments (3, 5 and 7% in each case). In figs. 6 and 7 the predictions of the TUBRNP model are compared with those of the Matsumura and Kameyama model for the burnup and radial shape function: as can be seen the agreement is excellent.

6. Summary

A new model for the nonuniform radial behaviour of the power density in high burnup fuel rods has been developed. The TUBRNP model is based on an extension of the RADAR model of Palmer et al. [2] and has been incorporated in the TRANSURANUS code [6]. Extensive comparison with local Pu measurements, local burnup data and with the purely empirical model of Matsumura and Kameyama have shown that confidence can be placed in TUBRNP predictions. In particular the radial shape function proposed fits closely the Pu radial concentration distribution measured in a variety of different fuels with varying enrichment (2.9 to 8.25%), burnup (21 000 to 64 000 MWd/t) and type.

Acknowledgements

The authors would like to thank G. Nicolaou for the KORIGEN calculations and M. Manolova for assistance with the manuscript.

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