A MODEL FOR THE DESCRIPTION OF THE EVOLUTION OF PU AGGLOMERATES IN MOX FUELS

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Abstract

In order to describe the irradiation behaviour of Pu agglomerates under LWR steady state conditions in MIMAS MOX fuels, a model including the neutronic evolution of the heavy atoms and their diffusion processes between the agglomerates and the matrix has been developed. It leads to the calculations of Pu enrichment in the two phases and of the agglomerates size evolution during irradiation. The calculated distribution of the fission in the fuel gives access to the local power and burnup heterogeneity factor. Electron probe microanalyses (EPMA) have been carried out on fuels irradiated up to 45000 MWd/tM. Diametral and local distribution of Pu are used to calculate the enrichments of the agglomerates and the matrix, which are then compared to the results of the model. During irradiation, the Pu concentration falls markedly in the agglomerates and increases steadily in the matrix, leading to a homogenization of the fuel on a microstructural scale. Heterogeneity factors give an estimate of the deviation from homogeneity. Knowing the local fission rate and burnup in the agglomerates and the matrix enables the calculation of local fission gas concentrations, which are compared to the xenon EPMA diametral distribution. Comparison with the calculated matrix xenon concentration at the edge of the pellet where there is no gas release, shows that some fission gas atoms which originated from the agglomerates, have been dissolved in the matrix by recoil. The calculated gas concentrations give an estimate of the quantity of gas dissolved. This work has been performed with the intent to improve fuel rod performance code estimates of fission gas concentrations retained or released in both the matrix and the agglomerates.

1. INTRODUCTION

In MIMAS (MIcronized MASter blend) MOX fuels, fabrication route induces an heterogeneous microstructure characterized by high Pu-content agglomerates - favouring high burnup values during irradiation - dispersed in a low Pu-content matrix.

Differences between these two phases are clearly revealed by post-irradiation examinations (PIE) and show a fission product accumulation, and an important porosity development in the agglomerates.

However, the fuel tends towards homogeneity as irradiation proceeds, due to heavy atom neutronic and diffusion processes, which favour the equalization of Pu in the matrix and in the agglomerates, and an implantation in the matrix of fission gas created in the agglomerates.

A heterogeneity model has been developed in order to (i) calculate and predict the fuel microstructure after base irradiation in order to compare the results with PIE, essentially electron-probe microanalyses (EPMA), (ii) estimate quantitatively the fission gas concentrations both in the agglomerates and the matrix for a coupling with fission gas release model of fuel rod performance code.

2. PU AGGLOMERATES EVOLUTION MECHANISMS

2.1 NEUTRONIC EVOLUTION

Neutron flux induces a neutronic evolution of the heavy atoms in the fuel. A simplified neutronic chain including U and Pu isotopes has been used (Fig. 1). One group cross-sections have been deduced from results obtained by the CEA 99 group neutronic code APOLLO, for a normal irradiation of a MOX fuel in a LWR. For each nuclide, an absorption cross-section (σ_a) and for fission atoms, a fission and a capture cross-section (σ_f and σ_c) are used. The evolution of the concentrations c_i can then be written as:

$$\frac{dc_i}{dt} = A_i - B_i$$

where A_i is the creation rate of the nuclide (worth 0 for U235, U238 and Pu238)$_i$ and B is the disappearance rate.

For example, the A and B terms for Pu241, deduced from the neutronic chain are:

$$A_{Pu241} = \sigma_{aPu240}\, c_{Pu240}\, \Phi$$

$$B_{PU241} = \sigma_{aPu241}\, c_{Pu241}\, \Phi - \lambda_{Pu241}\, c_{Pu241}$$

where σ_{aPu240}, σ_{aPu241} are the absorption cross-sections for Pu240 and Pu241, Φ is the neutron flux and $\lambda_{Pu241} = \ln(2)/T_{pu241}$, T_{Pu241} for the Pu241 period.

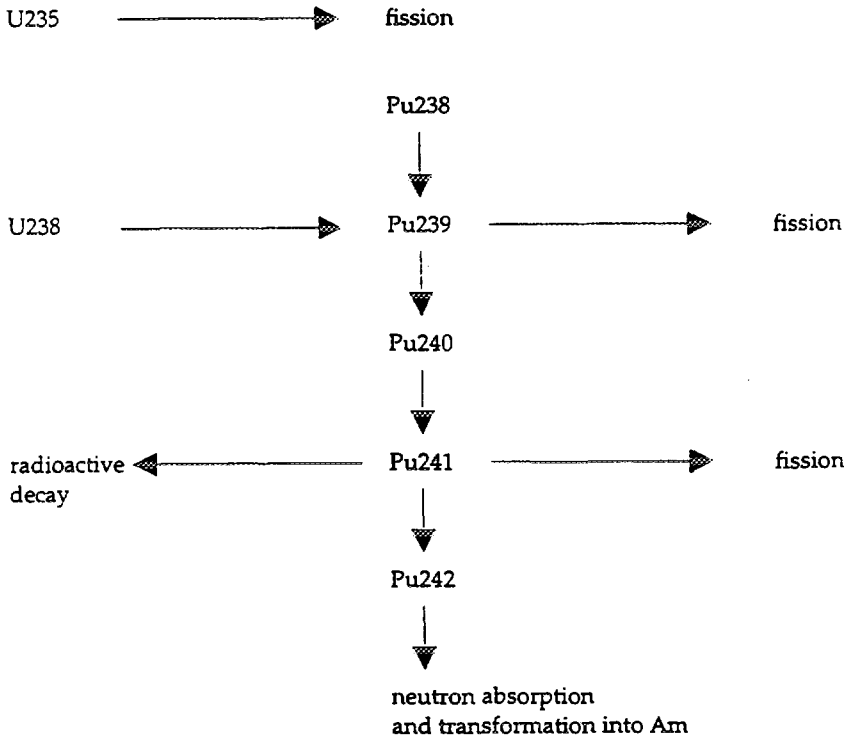

FIG. 1 Neutronic chain used in the heterogeneity model

2.2 DIFFUSION

Thermal and irradiation-enhanced metal diffusion of U and Pu occur during irradiation. For normal conditions, fuel temperature does not exceed 1300°C. Below this temperature, the diffusion coefficient is athermal and doesn't depend on Pu content. It can be described by:

$$D = k\hat{F}$$

where $\hat{F}$ is the fission rate (fissions. cm^{-3}. s^{-1}), and k is a constant equal to 1.2 10^{-29} cm^5 [1].

Note that for off-normal conditions, thermal diffusion could be added without major modifications.

The agglomerates are considered as spherical. Concentrations depend on radius (r), the centre of the agglomerate being chosen as the origin of coordinates (r = 0). Diffusion equation in spherical coordinates is used:

$$\frac{\delta c_i}{\delta t} = \frac{1}{r^2}\frac{\delta}{\delta r}\left(r^2 D \frac{\delta c_i}{\delta r}\right)$$

2.3 COUPLING NEUTRONIC AND DIFFUSION

The change with respect to time of concentrations is the sum of the neutronic and diffusion evolution defined in the last two sections. The equations are coupled via the neutronic terms which mix the concentrations, and the system to be resolved is non linear, due to the fact that the diffusion coefficient depend on the fission atom concentrations.

The numerical resolution is performed as follows: knowing the heavy atoms concentrations are function of radius at time step t_n, one wishes to calculate them at time step t_{n+1}. First, the neutron flux Φ is calculated from the power history of the rod, then the diffusivity is calculated according to:

$$D_{n+1} = k\Phi(\sigma_{fU235}c^n_{U235} + \sigma_{fPu239}c^n_{Pu239} + \sigma_{fPu241}c^n_{Pu241}$$

The concentrations are then calculated using a finite difference resolution scheme.

3. COMPARISON WITH EXPERIMENTAL RESULTS

3.1 FUEL CHARACTERISTICS

Fuel rods used for comparison with model calculations are part of the Saint-Laurent-B1 program [2]. A total of 16 rods fabricated using the MIMAS process have been thoroughly examined in hot cells in CEA Saclay and CEA Cadarache with careful attention being given to the fuel microstructure obtained by EPMA.

The MIMAS process is known to produce a heterogeneous distribution of Pu in agglomerates on a microscopic scale. Their Pu content is close to the master-mix enrichment (# 30 wt% Pu), and they are dispersed in a low-Pu content matrix [3].

Three mean pellet plutonium contents are used within an assembly: 2.88, 4.36 and 5.57 wt% Pu in the present case. Typical average agglomerate diameter, as measured by alpha-autoradiography, is close to 20 μm.

The heterogeneity model is applied to a class of agglomerates defined by its average radius and average enrichment, dispersed in a Pu-free matrix. With this hypothesis, the initial agglomerate volumetric fraction is:

$$F_V = \frac{e_{pell.}}{e_{aggl.}}$$

where $e_{pell.}$ is the average pellet Pu enrichment and $e_{aggl.}$ is the agglomerate Pu content. For the three zones, F_V = 9.6 (low Pu-content), 14.5 (medium Pu-content) and 18.6 (high Pu-content) vol%.

Fuel rods have been extracted after one, two and three irradiation cycles. Their highest mean (resp. peak) burnup is 41000 (resp. 46500) MWd/tM, and their linear power falls in the range 150 to 220 W/cm.

3.2 HEAVY ATOM EVOLUTION

Typical calculated evolution of Pu concentration in the agglomerates and in the matrix is shown in figure 2 after one, two and three irradiation cycles.

Neutronic evolution leads to a decrease in Pu concentration in the agglomerates and an increase in the matrix. Their value tends towards the mean Pu concentration, which can be calculated supposing the fuel to be homogeneous. Slight diffusion of Pu in the matrix occurs on a length lower than 5 μm.

Deviation from homogeneity can be estimated by heterogeneity factors. The power (resp. burnup) heterogeneity factor is defined as the ratio of the agglomerates or matrix power (resp. burn-up) versus their mean pellet value. Their initial value is close to the ratio of initial Pu concentration in the agglomerate or the matrix against mean Pu enrichment, i. e. nearly null for the matrix and 10.4 (low Pu content), 6.9 (medium Pu content) and 5.4 (high Pu content) for the agglomerates. They tend towards 1 as the irradiation proceeds. An example of their evolution versus burnup is shown in figure 3.

These results have been checked against EPMA, which enables the measurement of Pu concentration in the agglomerates and the matrix. Local analyses with a resolution of 2 μm clearly show

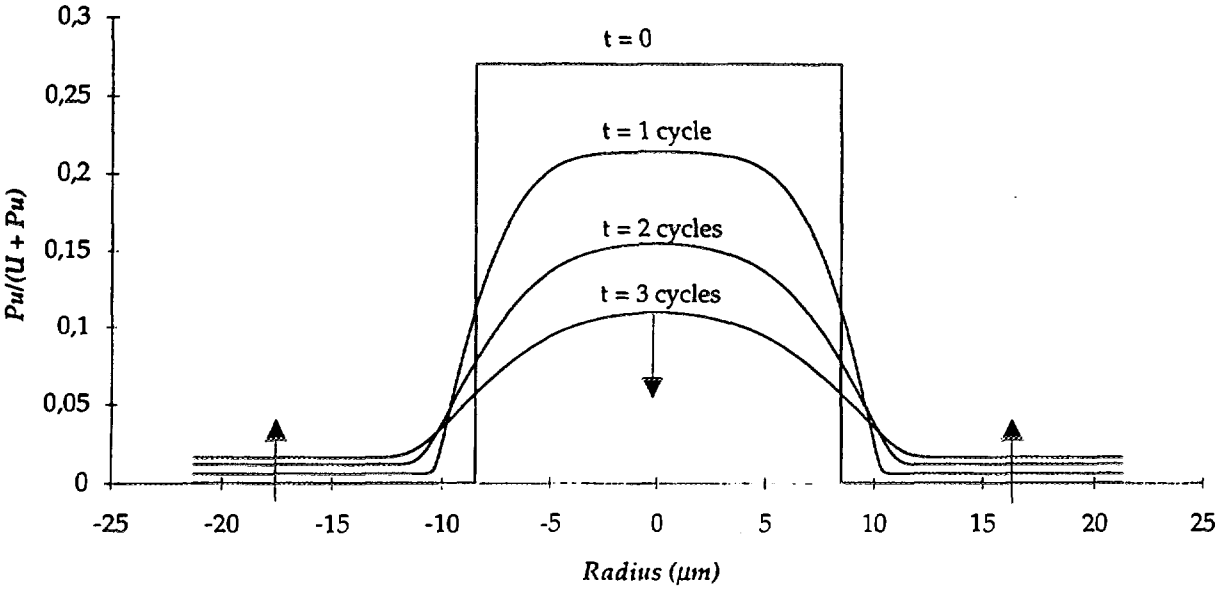

FIG. 2 Pu profile evolution during irradiation

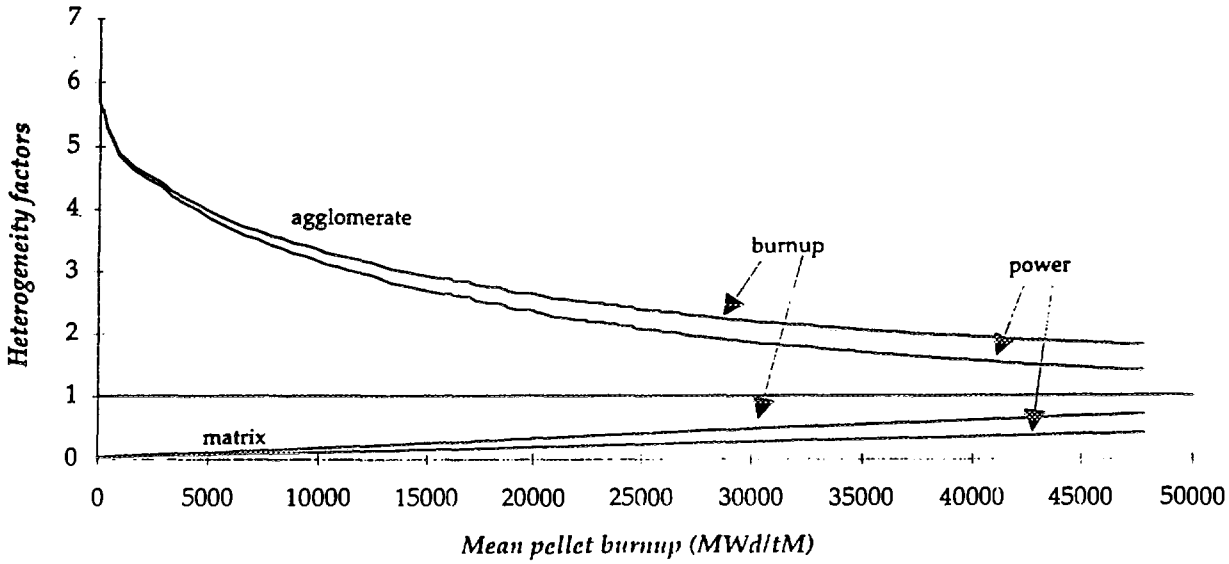

FIG. 3 Power and burnup heterogeneity factors of agglomerates and matrix versus mean pellet burnup

the agglomerate structure and particularly the Pu enrichment; diametral measurements enable the calculation of the matrix content. They are shown in figure 4 together with model calculations for the intermediate Pu-content zone.

3.3 FISSION GAS REPARTITION

Fission gases are created for a large part in the agglomerates where most of the fissions occur. The rates of birth of Xe and Kr for MOX fuels under normal conditions have been calculated using the APOLLO code and are used with the heterogeneity factors to calculate the gas distribution between the agglomerates and the matrix.

Another important phenomenon is taken into account: a significant amount of the gas atoms created within the agglomerates is implanted in the surrounding matrix by recoil. Gas atoms which are concerned must have been created in the range of fission fragment (i. e. of the order of 10 μm) from the agglomerate surface. A parameter named f_d, representing the fraction of fission gas created in the agglomerates and dissolved in the matrix has been introduced. The expressions for the gas concentrations are then:

$$GC_a^{aggl.} = GC^{aggl.}(1 - f_d)$$

$$GC_a^{mat.} = GC^{mat.} + GC^{aggl.} f_d \frac{x_{aggl.}}{1 - x_{aggl.}}$$

where $GC_a^{aggl.}, GC_a^{mat}$ (resp $GC^{aggl.}$ $GC^{mat.}$) are the created fission gas concentrations after (resp. before) implantation in the matrix and $x_{aggl.}$ is the agglomerate volumetric fraction.

Note that:

$$x_{aggl.} GC_a^{aggl.} + (1 - x_{aggl.}) GC_a^{mat.} = x_{aggl.} GC^{aggl.} + (1 - x_{aggl.}) GC^{mat.} = GC^{pell}$$

where GC^{pell} is the average pellet fission gas concentration.

This approach could also be used in case of an initial distribution of agglomerate diameter, to take into account the as-fabricated fuel microstructure. The f_d factor should then be written to show explicitly its proportionality with the free surface of agglomerates.

The diametral Xe EPMA of fuel rod pellets from the three Pu content zones, various power histories and burnups have been used to adjust the f_d parameter. A value of 25 % has been found to reproduce correctly xenon measurements in the matrix near the edges of the pellet where no fission gas release occurred (Fig 5, 6 and 7). As the burnup increases, fission gases remaining in the agglomerates accumulate in bubbles and are less visible with EPMA.

The value obtained for f_d is slightly lower but close to the one calculated by Walker and al. [4] (33 %) on MOX fuels whose fabrication process is similar to MIMAS.

In figure 7, thermal release of Xe from the matrix is seen at the centre of the pellet, due to a high power level reached by the rod on its third irradiation cycle (# 220 W/cm).

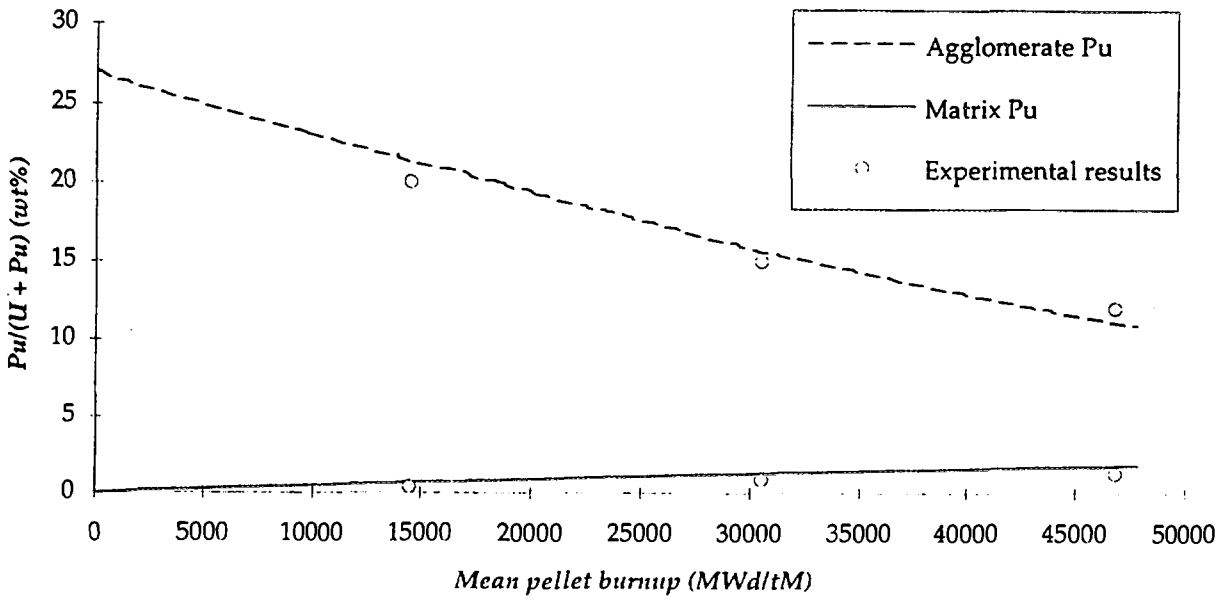

FIG. 4 Calculated andmeasured Pu content in the agglomerates and the matrix

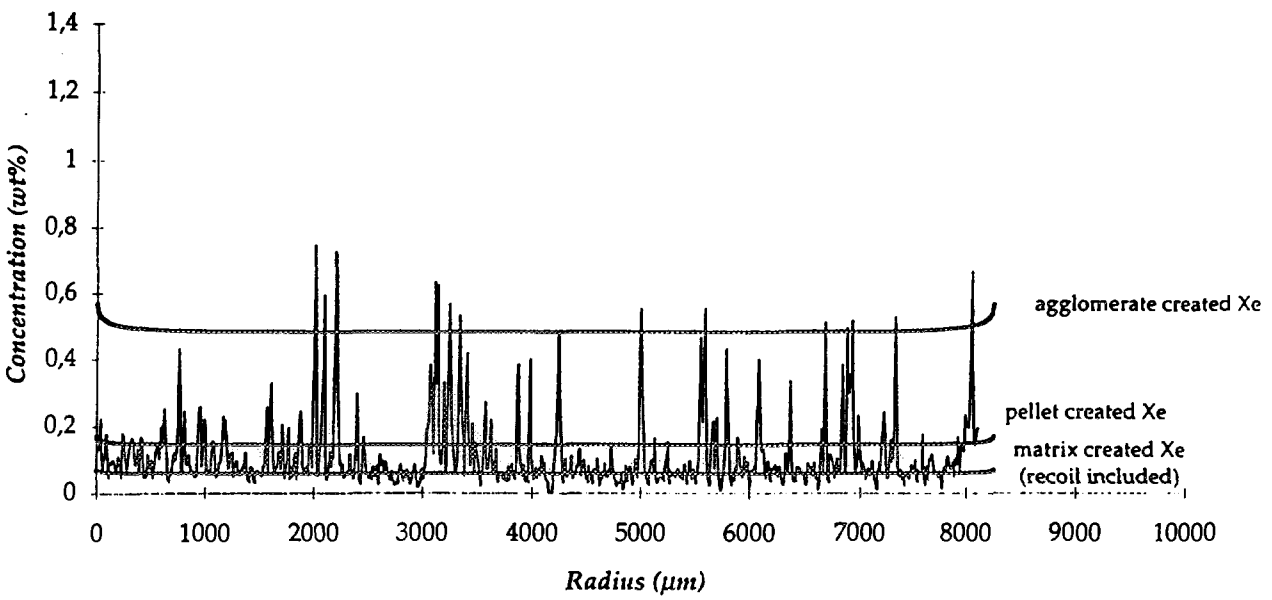

FIG. 5 Xenon diametral distribution - I irradiation cycle

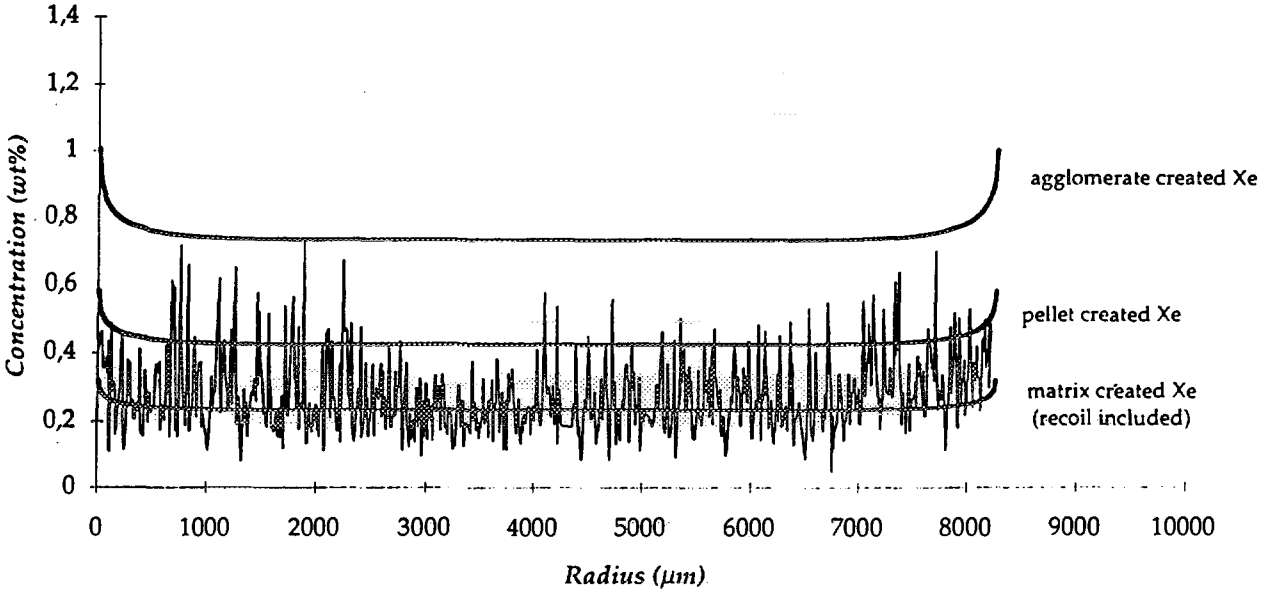

FIG. 6 Xenon diametral distribution - 2 irradiation cycles

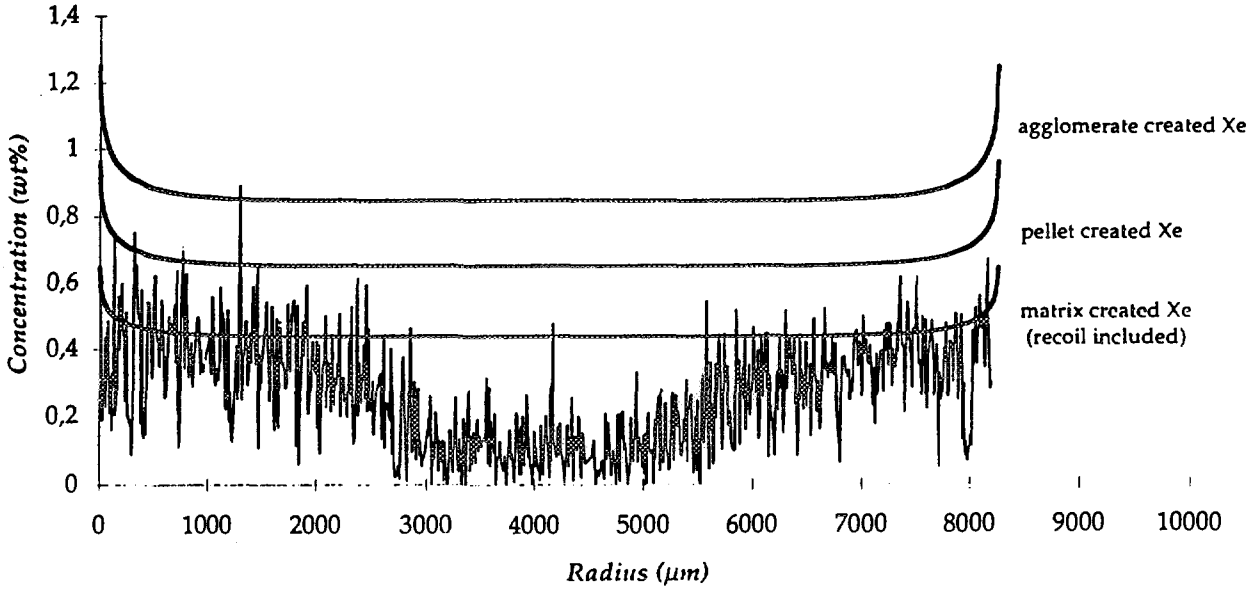

FIG. 7 Xenon diametral distribution - 3 irradiation cycles

4. CONCLUSION

MIMAS MOX fuels could be qualified as "moderately heterogeneous". Indeed, the Pu enrichment of the as-fabricated fuel is limited by the master-mix step to about 30 wt% Pu and the irradiation tends to homogenize the fuel microstructure mainly through neutronic processes, which overcome diffusion in normal conditions, and by a decrease of fission gas overconcentrations in the agglomerates due to recoil in the matrix.

The developed heterogeneity model yields quantitative estimates of the heavy atom and fission gas concentrations both in the agglomerates and the matrix.

Complementary results for as-fabricated MOX fuels are underway at Cadarache and should give more information on size and Pu enrichment distribution in the agglomerates. High burn-up effects in the agglomerates (fission gas release, precipitation of metallic fission products, ...) should also be included in the model whose results should confirm and explain the behaviour of MIMAS MOX fuels in normal conditions. The concentrations calculated by the model should also be used as initial conditions for fuel behaviour studies under off-normal conditions.

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