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FINITE ELEMENT ANALYSIS OF LOCAL OVERHEATING WITHIN PLUTONIUM ENRICHED UO_2 FUEL RODS CAUSED BY PuO_2 ISLANDS *

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Within natural UO_2 fuel elements enriched with plutonium, this last material should form PuO_2 solid solutions inside the UO_2 pellets, in a wide range of concentrations. If the solutions are obtained by mechanical mixing of the oxides, PuO_2 islands are formed in the UO_2 matrix. These islands may be the source of several problems in the fuel behaviour, the most important being the overheating of the matrix in the neighbourhood of the particles. It is caused by the large fission cross section of plutonium compared with that of uranium.

A detailed study of the thermal effects produced by PuO_2 particles in the UO_2 matrix and the cladding is then important for the specification of their permissible size. A portion of the fuel rods with spherical particles in the most significant places was studied. In order to obtain the dimensionless overheating of the fuel and cladding produced by the presence of those particles, the spacial distribution of temperature was calculated, solving the stationary and linear bidimensional equation of heat conducting using a finite element code. Several geometrical variables and material properties have been taken as dimensionless parameters. A satisfactory convergence of the numerical results to an asymptotic limit with a well-known exact solution, has been obtained.

1. Introduction

In the next few years, plutonium recycling in thermal reactors of uranium dioxide moderated with heavy water is economically promising. Much effort is therefore being made in several countries to develop the manufacturing technology of UO_2 fuel elements enriched with plutonium, and to study its behaviour and performance under in-reactor conditions.

Within these fuel elements, the plutonium should form PuO_2 solid solution inside the UO_2 pellets, in a wide range of concentrations and at the temperatures that these pellets reach in the reactor. The relevant physical properties of these solutions that affect the fuel elements' behaviour (such as thermal conductivity, melting points, mechanical properties, etc.) are similar to those of natural UO_2 , especially at the low

concentrations which may be used.

Amongst the procedures for manufacturing these solid solutions the mechanical mixing of the oxides takes a relevant place. In spite of its advantages, this procedure presents some problems because the low coefficients of cationic diffusion do not permit the homogenization of the mixture at normal temperatures and durations in the sintering processes [3].

Such principal inconveniences are:

- (a) Of manufacturing: to reach a level of mixing and sintering that ensures the required density and continuity at the PuO_2-UO_2 interfaces;
- (b) Of behaviour: The islands of the PuO_2 phase in the UO_2 matrix can produce:
 - (1) Effects of selfshielding;
 - (2) Adverse effects in the dopler coefficient in transients;
 - (3) Points of overheating in the matrix.

A detailed study of these physical effects caused by the PuO_2 particles in the UO_2 matrix is therefore important for the specification of their permissible size. In particular, item (3) is of great importance and

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its quantitative analysis is the objective of the present paper.

For specific cases, there are two factors that limit the permissible increment of the temperature in the fuel and in the cladding:

(a) on the symmetry axis of the pellets the temperature must not reach the melting point of the fuel material; and

(b) in the clad, alterations of its mechanical properties and acceleration of the susceptibility to corrosion must be avoided.

Knowing the relations between the size of the particles and the overheating that they produce in critical zones of the rods, for each of the two aspects (a) and (b) the corresponding maximum sizes can be calculated. For the least size, and for a given concentration of the solution, one must calculate the probability that two islands may be contiguous. If this value lies below that of the permissible limits of confidence, one must choose it as the value of specification of the island size.

A portion of the fuel rod with spherical particles of PuO_2 in the most significant places was studied. In terms of dimensionless parameters the spatial variation of the temperature in those geometries was obtained numerically using the finite element method. In order to determine the dimensionless overheating of the matrix produced by these particles, it is necessary to make a linear approximation of the problem, i.e., to use thermal conductivities not depending on the temperature. This approximation has practically no effect upon the values of the mentioned overheating [3].

2. Formulation of the problem

The present problem of spatial distribution of temperature is essentially tridimensional, but from both the points (a) and (b) of the introduction it can be seen that there are two fundamental situations to be considered:

(i) The particle lies on the symmetry axis of the pellets; and

(ii) It is contiguous to or near the lateral surface of the pellet beside the cladding.

In this paper we will consider only spherical particles. For the first case, the geometry is strictly

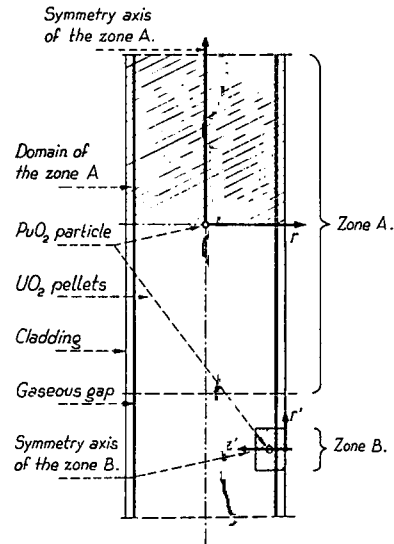


Fig. 1. Geometry of the problem under study.

axisymmetric (see fig. 1, zone A). Also, because the size of interest for the particles (not more than 500 microns) is very much smaller than the typical dimensions of the fuel rods, it is a very good approximation to solve the problem for case (ii) in a small portion of the pellet beside the cladding and containing it, and to approximate the actual geometry by one of axial symmetry, with the axis perpendicular to the surface of the cladding (see fig. 1, zone B) and passing through the centre of the particle.

It is clear that a complete tridimensional analysis of the problem for arbitrary position of the islands would not yield more substantial information than that obtained by the cited bidimensional axisymmetric approaches; further support for this argument will be given in the analysis of the results. For both situations the problem is then governed by the linear steady-state heat conduction equation, which in cylindrical coordinates (r, z) for axisymmetrical geometries, takes the form:

$$\frac{\partial}{\partial r} \left(K \frac{\partial T}{\partial r} \right) + \frac{\partial}{\partial z} \left(K \frac{\partial T}{\partial z} \right) + \frac{K}{r} \cdot \frac{\partial T}{\partial r} - \rho(r, z) = 0, \quad (1)$$

where $T = T(r, z)$ is the temperature at each point; $K = K(r, z)$ is the thermal conductivity (different for each material) which we consider independent of temperature in order to obtain dimensionless results; and $\rho(r, z)$ is the heat generated per unit volume.

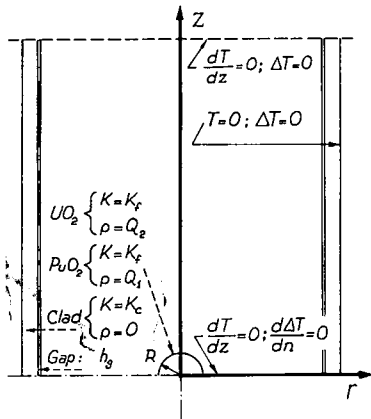


Fig. 2. Data for zone A.

which is Q_1 in the PuO_2 sphere, $Q_2 \ll Q_1$ in all the rest of the pellet, and zero in the cladding and in the gaseous gap. For simplicity we suppose equal conductivity for both the PuO_2 and UO_2 , because there is a little difference between them. For both the zones of interest, indicated in fig. 1, the domains where (1) must be solved are shown in figs. 2 and 3 respectively, in which the variable dimensions and magnitudes that enter in the analysis and all the boundary conditions are defined.

Because we are only interested in the distribution of the increment of temperature ΔT that the presence of the particles originates, we solve (1) directly for this increment $\Delta T(r, z)$, with $\rho = Q_1 - Q_2$ on the PuO_2 sphere and $\rho = 0$ in all the rest of the domain.

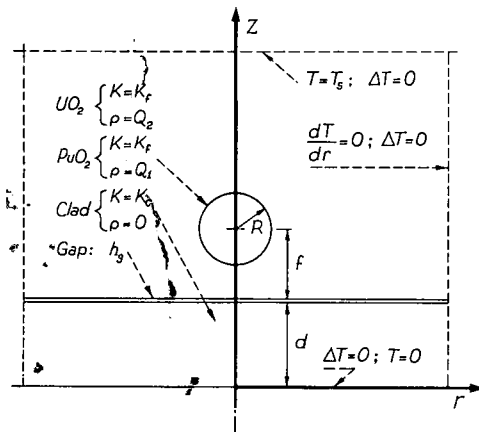


Fig. 3. Data for zone B.

In this case we must impose the condition $\Delta T = 0$ on all the boundaries. The latter is valid because of the linearity of eq. (1).

3. Numerical solution by the finite element method

The problem was solved using a finite element code, named CUARM [1] for bidimensional heat transfer problems. Toroidal trinoidal elements with triangular sections and first degree polynomials for the variation of the temperature within these element were used. The resulting linear system of equations is solved by the Gauss elimination algorithm for band matrix. The element meshes for both the domains were constructed automatically, for several configurations with specific sets of values for the geometrical parameters. Examples of both types of meshes are shown in figs. 4 and 5 respectively.

Results are obtained in terms of the dimensionless magnitude $\Delta T / [Q_2(F - 1)R^2/K_f]$, where F equals Q_1/Q_2 , K_f is the conductivity of the fuel, and R is the radius of the PuO_2 sphere. F can be calculated for a

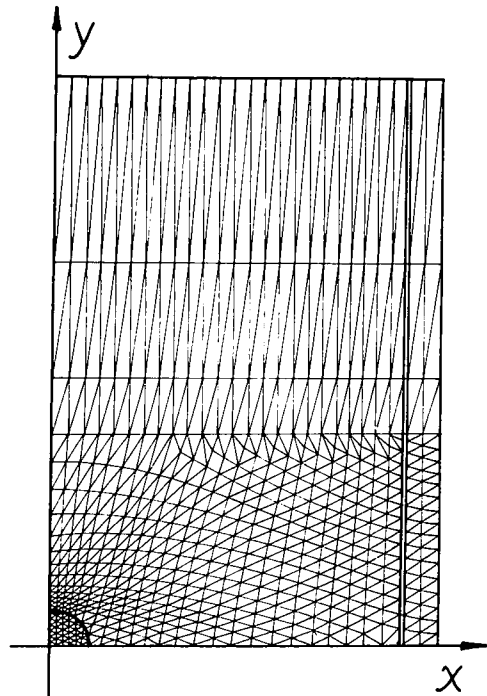


Fig. 4. Typical finite element mesh for zone A.

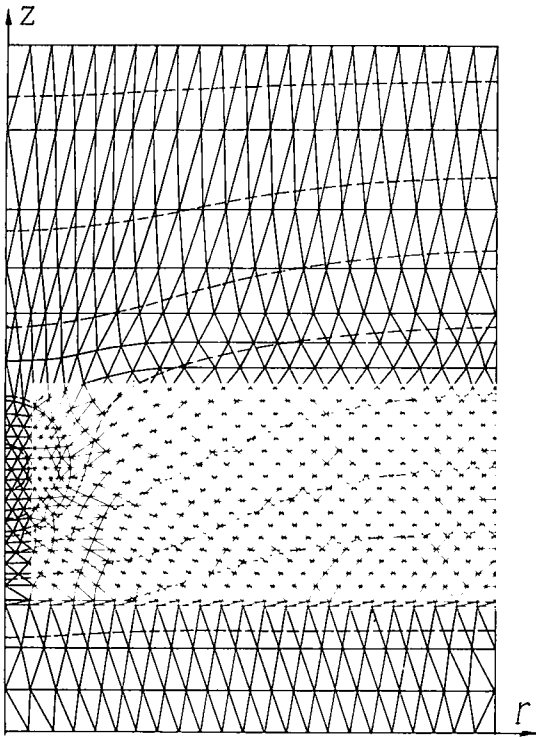


Fig. 5. Typical finite element mesh for zone B.

specific case in terms of the concentrations of the ²³⁹Pu and ²⁴¹Pu isotopes in the PuO₂, as indicated in Appendix A.

For the case of fig. 2, the dimensionless increment of temperature in both the centre of the sphere and in the sphere-matrix interface are shown in fig. 6, plotted versus the ratio between the pellets and the PuO₂ sphere radii. The following magnitude has been taken as the parameters of the curves:

$$\xi = \frac{h_T}{K_f/R_p} = \frac{R_p/K_f}{d/K_c + 1/h_g}, \quad (2)$$

where R_p is the radius of the pellet, d the thickness of the cladding, K_c the conductivity of the cladding material, h_g the conductance of the fuel-cladding gap, and h_T the total conductance of this gap and the cladding together. ξ represents the influence of the last two factors on the total heat transfer in the system under consideration.

The dimensionless maximum increments of tem-

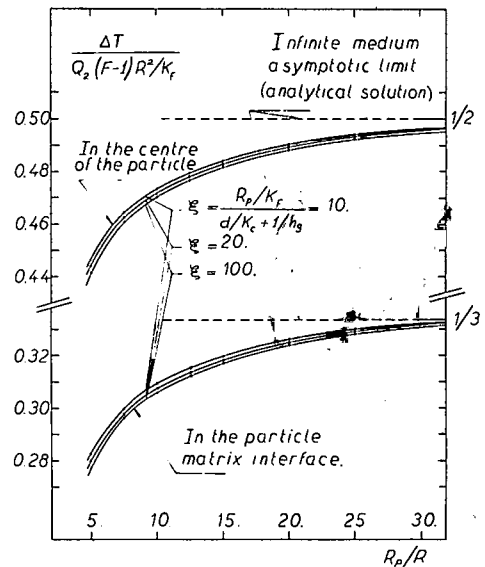


Fig. 6. Dimensionless increment of temperature for the particle centred on the symmetry axis of the rod (zone A).

perature within the particle and in the particle-matrix interface for the case of fig. 3 have been calculated in terms of the depth f of this particle within the pellet, related to its radius. In fig. 7 we represent these results. Now, the magnitude

$$\eta = \frac{h_T}{K_f/f} = \frac{f/K_f}{d/K_c + 1/h_g}, \quad (3)$$

which plays a similar roll to that of ξ , is the parameter of the curves. Similarly to the former case, in fig. 5 the isotherms for a specific set of parameters are shown.

Finally, for reason (b) mentioned in the introduction, it is important to know the maximum increment of temperature on the internal surface of the cladding just in front of the PuO₂ particle. That magnitude is plotted in fig. 8, also against the ratio f/R , with the same parameter η , and depending on the relation between the cladding and the gap conductance.

It is important to point out that plotting this information only once, for a general value of $(K_c/d)/h_g$, is an approximation which results from the assumption that the heat flux in the zone of the cladding in front of the particle is strictly perpendicular to its surfaces, i.e., not depending on the r -coor-

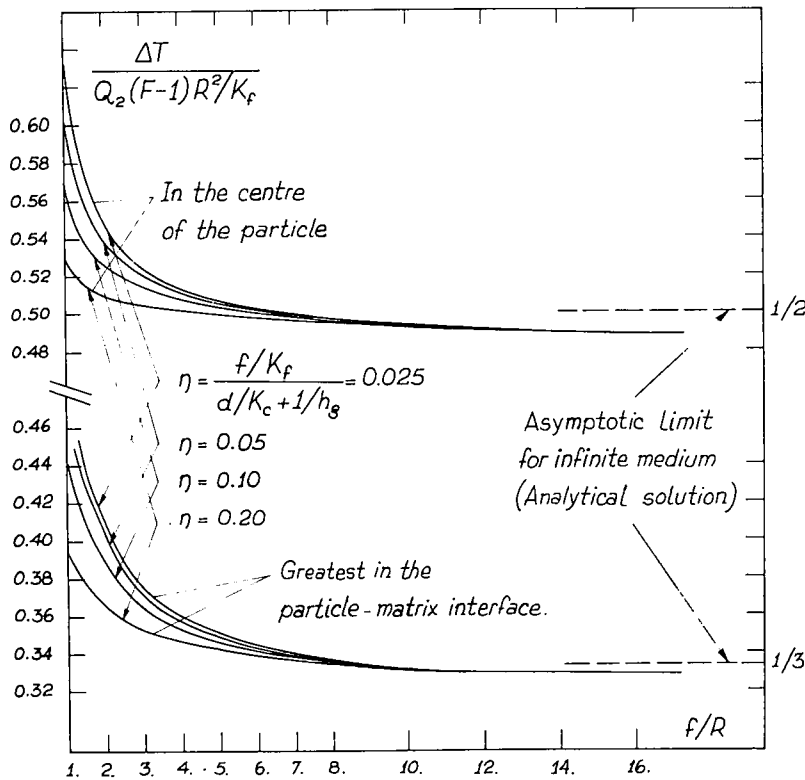


Fig. 7. Dimensionless increment of temperature for the particle near the cladding (zone B).

dinate in fig. 3. It was calculated that for a range of practical values of the ratio $(K_c/d)/h_g$ (from 20.0 to 4.0) maintaining η constant, the ordinates in fig. 8 vary by only 4% for $f/R = 1$, and this difference

decreases for greater values of f/R . Thus, this approximation is satisfactory for practical purposes.

4. The asymptotic limit for infinite medium

It is clear that the spatial variation of the increment of temperature for the two situations treated, must converge asymptotically to the case of an homogeneous sphere in an infinite medium of the same thermal properties. This asymptotic limit has an elementary exact analytical solution.

Indeed, let us consider a sphere of radius R of thermal conductivity K within which there is a uniform heat generation per unit volume Q_1 , immersed in an infinite medium of the same thermal conductivity and uniform heat generation Q_2 . The value of the temperature obviously diverges, but the increment ΔT of the temperature caused by the particles must satisfy the steady state heat conduction equation in spherical coordinates for systems of radial

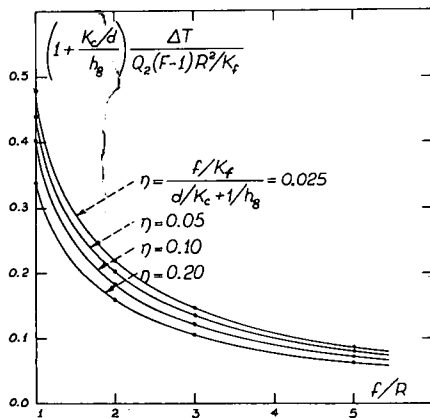


Fig. 8. Maximum increment of temperature on the internal surface of the cladding just in front of the particle.

symmetry:

In the PuO₂:

$$\frac{d^2 \Delta T_1}{dr^2} + \frac{2}{r} \frac{d\Delta T_1}{dr} + \frac{Q_1 - Q_2}{K} = 0. \quad (4)$$

In the UO₂:

$$\frac{d^2 \Delta T_2}{dr^2} + \frac{2}{r} \frac{d\Delta T_2}{dr} = 0,$$

with the conditions of continuity in temperature and heat flux in the interface of the two media, finite at the centre of the sphere, and zero at infinity. Solving system (4) with those boundary conditions in the standard analytical way, we arrive at the well-known solution:

In the PuO₂:

$$\Delta T_1(r) = \frac{Q_1 - Q_2}{6K} (3R^2 - r^2). \quad (5)$$

In the UO₂:

$$\Delta T_2(r) = \frac{Q_1 - Q_2}{3K} \frac{R^3}{r}.$$

Thus, the asymptotic limits to which the numerical solutions of $\Delta T/[Q_2(F-1)R^2/K_f]$ must converge in the centre of the sphere and in the sphere-matrix interface from (5) are $\frac{1}{2}$ and $\frac{1}{3}$ respectively.

5. Discussion of the results and conclusions

The satisfactory convergence of the numerical results to the exact analytical solution for the limiting case, that can be seen in figs. 6 and 7, gives reliability to the obtained plots.

For practical sizes of PuO₂ particles located on the symmetry axis of the pellets, the influence of the fuel rod's cooling on the overheating studied is not large, as we can see in fig. 6. Additionally, in fig. 7 we note that the same fact occurs for particles near the cladding for which $f/R \geq 4$. Then, in order to obtain a design criterion we can consider valid the results of fig. 6 for an arbitrary place within the pellets far enough from the cladding. These results also corroborate the assertion previously given that a tridimensional analysis of this problem is unnecessary for the particle sizes that occur in practice.

Since the results of figs. 6 and 8 have been expressed in dimensionless units, these graphs provide general information for the designer of any kind of mixed oxide fuel elements with micro-heterogeneities, enabling him to solve problems concerned with safety aspects of such fuels.

Appendix A. Calculation of the F ratio

Let properties corresponding to the PuO₂ and UO₂ be represented by variables with suffices 1 and 2 respectively. Assuming that the presence of the PuO₂ particle does not affect the position of the thermal neutron flux $\phi_{th}(r)$ within the matrix, the ratio F between the heat generated in each material is:

$$F = \frac{Q_1}{Q_2} = \frac{\Sigma_1^f \phi_{th}(r)}{\Sigma_2^f \phi_{th}(r)} = \frac{(\delta_1/A_1) \sigma_1^f}{(\delta_2/A_2) \sigma_2^f},$$

where Σ_1^f and Σ_2^f are the macroscopic fission cross-sections for thermal neutrons; σ_1^f and σ_2^f their corresponding microscopic cross-sections; A_1 and A_2 the molecular weights; and σ_1, σ_2 the densities, all for both materials.

If C_{239} and C_{241} are the normalized concentrations of the ²³⁹Pu and ²⁴¹Pu isotopes in the PuO₂ (²⁴⁰Pu does not have significant fission cross-section), taking standard values [2,4] for the physical magnitudes, we obtain

$$\begin{aligned} F &= \frac{11.46 \cdot 270(742.5C_{239} + 1000.9C_{241})}{10.96 \cdot 274 \cdot 0.0072 \cdot 582.2} \\ &= 182.5C_{239} + 246.02C_{241}. \end{aligned}$$

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