INFLUENCE OF COATED PARTICLE STRUCTURE IN THERMAL NEUTRON SPECTRUM ENERGY RANGE

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COATED PARTICLE STRUCTURE

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Abstract

The heterogenity due to lumping the fuel in coated particles affects the thermal neutron spectrum. A calculational model is discussed which, apart from some simplifying assumptions about the statistical distribution, allows a rigorous computation of effective cross sections for all nuclides of the heterogeneous medium. It is based on an exact computation of the neutron penetration probability through coating and kernel. The model is incorporated in a THERMOS-code providing a double heterogeneous cell calculation, which can be repeated automatically at different time steps in the depletion code system MAFIA-V.S.O.P..

A discussion of the effects of the coated particle structure is given by a comparison of calculations for heterogeneous and homogeneous fuel zones in pebble bed reactor elements. This is performed for enriched UO_2 fuel and for a ThO_2-PuO_2 mixture in the grains. Depending on the energy dependent total sigmas in the kernels the changes of the cross sections are ranging from o.1% up to 45%. The influence on the spectrum averaged sigmas of the nuclides in the fresh UO_2 fuel is lower than 1%. For the emerging ^{240}Pu it increases up to 3.3% during irradiation. For the ThO_2-PuO_2 fuel the averaged sigmas of the isotopes vary from o.5% to 5.7% depending on the state of irradiation. Correspondingly there is an influence on the plutonium isotopic composition, on breeding ratios, and on the tilt of k_{eff} during burnup which will be discussed in detail.

Introduction

In gas cooled and graphite moderated reactors the fuel is actually lumped in little spheres which are overcoated by pyrolytic graphite. These coated particles are inbedded in a matrix of moderating material (e.g. carbon), and lumps of this mixture are surrounded by pure graphite forming the fuel element units for the HTR's. For the calculation of neutron flux and reaction rates a correct treatment of the double heterogenity is important in some ranges of the energy spectrum.

The reduction of the resonance integrals has been investigated by several authors assuming naked kernels (1,2) or overcoated particles (3,4) randomly distributed in the matrix. In the range of the thermal energy groups selfshielding factors for a heterogeneous grain structure have been derived by some authors. In order to get analytic expressions, several approximations are used. The assumptions of Dyos and Pomeraning (5) neglect the mutual shielding of the kernels, and Ragan's (6) one group model suppresses the different importance of the shielding for the various nuclides. The methods of Doub (7) and Askew and Carpenter (8) are restricted to naked kernels in a moderator. They neglect, that a neutron may traverse many coating regions of the particles even if the kernels are quite black. This holds also for the model of Lane, Nordheim and Sampson (1) which has been used as a fundament for double heterogeneous thermal cell calculations by George, Jaye and Astley (9).

The aim of this work is to develop a more accurate double heterogeneous cell calculation and to discuss the effects on spectrum averaged cross sections, on reactivity and on the change of the isotopic composition during burnup. The calculational model respects the mutual shielding of the coated particles which influences all reaction rates in kernel, coating and matrix material. It is based on the accurate

numerical computation of the penetration probability for a neutron traversing the coated particles, as derived in refs. (3,4). The model is used to define effective cross sections for all materials in the fuel region.

The computation of the effective cross sections is incorporated in a 30 group THERMOS version (10) which can be used for spectrum calculation in the depletion code system MAFIA-VSOP(11). The double heterogeneous cell calculation is automatically repeated at several time steps during burnup history of a reactor providing thermal spectrum averaged cross sections for the diffusion and depletion calculations.

Burnup calculations have been performed for two pebble bed fuel compositions (UO_2 and ThO_2-PuO_2). In order to discuss the grain structure effect these calculations are done both for a coated particle structure and for a homogenized medium in the fuel zone. The comparison will demonstrate the effect of the heterogeneous structure on some characteristic data. For the UO_2-fuel the grain structure effect changes the content of ^{240}Pu by 2% after a burnup of 58 800 MWd/t. During the irradiation period the discrepancy in the averaged absorption cross section of ^{240}Pu due to heterogenity raises up to 3.3% whereas it stays below 1% for all other nuclides. For the PuO_2-ThO_2 fuel the ^{240}Pu content differs by 3% already at 29 100 MWd/t. The effect on the cross sections of the different isotopes varies from 1.1% to 5.3% for the fresh fuel and from 0.5% to 5.7% for the irradiated one. An increase of this cross section shielding effect up to 60% can be achieved by using a mixture of coated particles in which the plutonium and the thorium are separated in different kernels. This can be of importance for advanced fuel designs.

The Calculational Model

In the coated particle structure medium the collision probabilities for a neutron are different from those in a homogenized mixture. This holds as well for the absorptions as for the scattering collisions in kernel, coating and matrix material. The idea is to define effective cross sections for each nuclide which enable to treat the fuel region as a homogenized "effective" medium.

The following assumptions are used:

1. On the path of a neutron through a statistical distribution of coated particles the mean value for the distances of subsequent geometrical cross sections is equal to $L = 1/Z\pi R^2$ (with Z = number of coated particles per cm^3, and πR^2 = cross section of one of them). For calculating the penetration probability through the matrix from one coated particle to the next one the model uses this mean L instead of averaging over any statistical distribution of the distances. The distribution of the geometric cross sections with respect to the path of flight is assumed to be random and neglects any correlation between the positions of neighbouring coated particles.

2. A source neutron born at energy E is assumed to start at a distance L/2 from the first coated particle cross section which lays in its flight direction, although the preceeding scattering may have occured in a matrix, coating or kernel material. Similarly for a neutron entering the heterogeneous zone the first cross section is assumed to be at a distance L/2 from the boundary. This assumption is sufficient if the mean free path through the heterogeneous medium is of the order of some coated particle distances L, which is true in normal realistic arrangements. For extreme cases a more general treatment is possible according to ref. 4.

A numerical method is used to compute the non collision probability P(E,X) for a neutron at energy E, which traverses a distance X in the heterogeneous medium. Then an effective total macroscopic cross section $\Sigma_T^*(E)$ can be defined by

$$P(E,X) = e^{-\Sigma_T^*(E)\cdot X} \qquad (1)$$

The effective cross sections for the particular reactions will be deduced by means of the collision probabilities in kernel, coating or matrix material.

Penetration Probability through one Coated Particle

The most general expression for the non collision probability in a sphere is given by

$$p = \int_{\substack{\vartheta=0..\frac{\pi}{2} \\ \varphi=0..2\pi}} \frac{\cos\vartheta}{\pi} \sin\vartheta \, d\vartheta \, d\varphi \, e^{-\int_{x=0}^{s(\vartheta)} \Sigma_t(x)\,dx}$$

This assumes an arbitrary position of the sphere in the flight direction of the neutron and a heterogeneous material distribution inside. The first term is the probability of hitting the sphere within $\sin\vartheta \, d\vartheta \, d\varphi$, and the exponential is the probability of traversing on the sinew $s(\vartheta) = 2R\cos\vartheta$ with a point dependent distribution of the total cross section. Assuming spherical symmetry for the total cross section $\Sigma_t(x)$ and $\cos\vartheta = u$ this becomes

$$p = \int_0^1 2u\,du\, e^{-\int_0^{2Ru} \Sigma_t(x)\,dx} \qquad (2)$$

In coated particles there are two spherical zones of different Σ_t, and p has to be evaluated numerically as derived in ref. 4. For a sphere with a homogenized Σ_h the probability is explicitely

$$p_1 = \frac{2}{(2R\Sigma_h)^2}\left[1-(1+2R\Sigma_h)e^{-2R\Sigma_h}\right].$$

An averaging of the exponent instead of the exponential leads to

$$p_2 = e^{-\int_0^1 2u\,du\cdot 2Ru\Sigma_h} = e^{-\frac{4}{3}R\Sigma_h}. \qquad (3)$$

which means that all neutrons are assumed to pass through on the mean cord length. This is what is actually done when homogenizing a grain structure medium. An expansion of p_1 and p_2 in terms of $R\Sigma_h$

$$\frac{p_1-p_2}{p_1} = \left(\frac{R\Sigma_h}{3}\right)^2\cdot\left(1+\frac{4}{45}R\Sigma_h+\dots\right)$$

shows that both of them are identical for $\frac{R\Sigma_h}{3} \ll 1$.

Fig. 1 gives a survey of the range of validity of the approximations for the penetration probability. Curve I is the p for a coated particle according to (2). It is given as function of the total microscopic cross section σ_t of the kernel. The data for the coated particle are: Inner radius o.o3 cm, outer radius o.o5 cm, inner material ^{238}U-metal with an atom density o.o45·1o^{2} cm^{-3}, outer material is graphite of o.o83·1o^{24} cm^{-3} and σ_{tot} = 4.7 barn. For large σ_t the kernel becomes black indeed, p, however, approaches a limit much greater than zero because of

the fraction of neutrons traversing the shell only. Curve II is p for a coated particle with the same number of atoms, but with a kernel of uranium carbide and corresponding an inner radius of o.o3466 cm. For curve III all materials inside the little sphere of the radius o.o5 cm are homogenized. Curve IV is the approximation p_2 of formula (3) which adequately represents the penetration probability in the homogenized medium.

Apparently an exact treatment of the coating shell becomes important as soon as σ_t of the kernels comes into the range of a few hundred barns per atom. Such cross sections are realistic in some intervals of the thermal spectrum, especially if plutonium is present. There is a chance for a neutron to pass through the range of some coated particles even if the kernels are real black. According to curve III this is not possible in a model which computes the grain structure shielding by means of the penetration probability through uncovered kernels (1,2,7,8,9).

In case a neutron interacts inside the coated particle

$$p_K = \int_0^1 2u\,du\, e^{-\Sigma_C S_C(u)} \cdot \left(1 - e^{-\Sigma_K S_K(u)}\right) \qquad (4)$$

is the probability for a collision in the kernel. Here $\Sigma_{C,K}$ and $S_{C,K}(u)$ are the total sigmas and the fractions of the sinews in coating and kernel, respectively. (For the numerical evaluation see ref. 4). The probability for a collision in the coating is

$$p_C = 1 - p - p_K \,. \qquad (5)$$

The ratios

$$R_K = \frac{p_K}{p_K + p_C} \quad , \qquad R_C = \frac{p_C}{p_K + p_C} \tag{6}$$

give the distribution of a collision on kernel and coating material, respectively. It should be noted that the numerical integrations for p and p_K need an accuracy of 8-1o digits because of the small differences in p_C.

In case of coated particles inbedded in a graphite matrix a collision may occure in this medium too. A neutron has to traverse a certain distance in this medium before hitting one coated particle and after leaving it. An accurate treatment should find the collision probability as an average over all possible starting points of the neutron. But because of so many uncertainties of the geometric arrangement the numerical computation assumes that a neutron beam traverses per coated particle a cylinder of the cross section πR^2 and the length L in axial direction as fig. 2 shows. In this L is the mean distance from one coated particle to the next one following the beam through the grain structure medium. This simplified model takes into account that neutrons stripping the coated particle have to traverse more matrix material than those hitting it in the center. Replacing a neutron by a homogeneous beam is equivalent to the assumption of a random position of the coated particle with respect to the flight direction of the neutron.

The probabilities p, p_K for penetrating or interacting in the kernel, respectively, can easily be defined analogously to the forms (2) and (4) on the basis of fig. 2(4). Automatically p_C in (5) becomes the probability for a collision in coating or matrix as well.

Effective Cross Sections for the Coated Particle Medium

For the use in computer codes it is convenient to simulate the grain structure by a homogeneous medium with effective cross sections. If there are z coated particle per unit volume a neutron enters n of them at a distance $X = n/z\pi R^2$. The probability for travelling this distance without any collision is p^n. This explicitely means the probability of traversing the areas of n subsequent coated particles without a collision which, apart from the assumption 1 for the calculational model, contains rigorously the "mutual shielding" of the grains. As mentioned above (eq.(1)) this can be used to define an effective macroscopic cross section Σ_T^* by

$$p^n = e^{-\Sigma_T^* X} \qquad \text{i.e.} \qquad \Sigma_T^* = -z\pi R^2 \ln p \,.$$

Assuming for p^n the approximation of (3) Σ_T^* actually becomes equal to the total sigma of the homogenized medium which is $\Sigma_T = \Sigma_h \cdot z \cdot \frac{4\pi}{3} R^3$.

By means of definition (6) the total cross sections for kernel or graphite collisions in this effective medium are

$$\Sigma_K^* = R_K \Sigma_T^* \,, \qquad \Sigma_C^* = R_C \Sigma_T^* \,.$$

The ratio Σ_K^*/Σ_K is the effective reduction of the kernel material cross sections compared to the homogenized medium. It is used to modify all cross sections and scattering matrices of all nuclides in the kernel. Correspondingly the efficiency of the graphite in coating and matrix is increased by Σ_C^*/Σ_C.

In order to show the range of importance of this effect fig. 3 presents this ratio for two different media in the energy range

from 0 to 2 eV (which means v = 0 ... 9 for the velocity in terms of 2200 m/sec). All curves hold for coated particles with inner radius 0.03 cm, outer R = 0.0[illegible] cm, $Z = 0.3 \cdot 3/4\pi R^3$, the density of coating and matrix is 1.65 and 1.6o gr/cm^3, respectively. The kernels of curve I consist of UO_2 with 8% enrichment of ^{235}U. The kernels of curve II contain a mixture of ThO_2 and PuO_2 with the Pu-isotopic composition 50/25/15/10 and an enrichment of 4% fissile Pu.

The effect is of importance in the lower energy range and in the Pu-resonances if present. It causes a change in the energy flux distribution and affects criticality and burnup behaviour of the reactor.

Concerning a variation of the coated particle sizes it has been shown in ref. 4 that for decreasing dimensions of the covered kernels the model automatically yields the sigmas of the homogenized medium, i.e. $\lim_{R \to 0} \Sigma^*_{K,C} / \Sigma_{K,C} = 1$. Here all results are derived only for the coated particle structure mentioned above and for the homogenized medium. The influence of size variation can be estimated from an interpolation.

In fig. 3 there is a dotted curve III far below the others. It holds for the same material composition and coated particle size as curve II. The only difference is that the ThO_2 is assumed to be mixed between the matrix material and only the plutonium is lumped into kernels of coated particles ($Z = 0.0185 \cdot 3/4\pi R^3$). There is a very strong shielding of the plutonium, and this effect encourages to extend the calculational methods to mixtures of two kinds of coated particles.

Coated Particle Shielding during Burnup

The numerical calculation of the effective cross sections has been incorporated in the THERMOS code, which gives a cell calculation on the basis of transport theory. THERMOS is a part of the new burnup program cycle MAFIA-V.S.O.P. (11) which automatically iterates on spectrum calculation at many burnup time steps. To give a survey about the coated particle shielding effect on reactor history we follow a simplified model of a pebble bed core by means of this program system.

The two coated particle structures described in the preceeding chapter are filled into fuel balls with an inner radius 1.5514 cm (1.4763 cm for the ThO_2-PuO_2 composition), outer radius of 3.0 cm. A reactor core with height and diameter of 7 m is made up of fuel balls at random distribution. The power density is 11 MW/m^3 and the average temperature 900°C. The depletion is not disturbed by shuffling and control poison adjustments. The burnup tendencies are symptomatic for a graphite moderated core with tubular fuel elements as well.

The core containing the 8% enriched UO_2 fuel (case I) comes to a burn up of 58800 MWd/t after a life of 800 days, the 4% enriched ThO_2-PuO_2 core (case II) reaches 29100 MWd/t after 400 days.

In fig. 3 the dotted curves I_b, II_b show the $\Sigma_\kappa^* / \Sigma_\kappa$ after this irradiation. In case I the shielding effect in the low energy range decreases and in the range of the Pu-resonances it increases. This is due to the fact that 82% of the initial ^{235}U has been lost in absorptions and plutonium has been built up. Overlapped is the effect by the fission products of which the ^{135}Xe decreases during burnup because of an increase in the absolute flux.
In case II ^{239}Pu and ^{240}Pu deplete, the ^{241}Pu content stays approximately the same, and after about 100 days of irradiation

the ^{233}U starts to build up rapidly because of ^{233}Pa-decay from Th-breeding. Due to the burn up of ^{239}Pu the cross section reduction at low energies should change similar as in the range of the ^{239}Pu-resonance at $v = 3.4$. But the bred ^{233}U and the fission products are creating an opposite tendency in that range of energy.

To show in detail the effect of cross section reduction on fuel history the burnup calculations are performed for the shielded grain structures and for elements with homogenized inner zones separately. The change in neutron spectrum and in the averaged quantities is somewhat different for the heterogeneous and homogeneous arrangements. In the heterogeneous cases materials with high cross sections in the energy ranges of high reduction are less important. If ^{240}Pu is present the production rate of ^{241}Pu is reduced.

Table I gives the decrease of k_{eff} during burnup. For the UO_2-coated particles the reactivity swing is less than in the homogeneous case because of the lower absorption in the ^{240}Pu which is produced during irradiation. For the ThO_2-PuO_2-core with particle structure the initial k_{eff} is higher than in the homogeneous case because of the ^{240}Pu grain shielding. The k_{eff}, however, decreases more rapidly due to the lower ^{241}Pu breeding gain.

The spectrum shift during burnup causes a change in the averaged cross section. The time dependency is not linear and varies for the different isotopes. In order to present only a survey of the change in the shielding effect the table gives the initial and final σ_a for a few isotopes. In the UO_2 core the effective reduction stays nearly constant about 0.5% for most of the nuclids. For ^{240}Pu, however, it increases from 0.8 to 3.3%. For the ThO_2-PuO_2 core there is a remarkable time dependency for all cross sections, and the effective shielding varies between 0.5 and 5.7%.

Correspondingly the breeding ratio and the composition of the Pu isotopes are influenced. Data are given in the table, too.

The very extreme case III with only few Pu-coated particles in a matrix consisting of all other materials is only a fictious one. A burnup calculation was not performed because there was no routine available to treat separate fission products from the Pu in the grains and from the ^{233}U in the matrix for the repeated spectrum calculation. The last column of the table shows, however, a very high reduction of Pu-cross sections for the initial loading. This effect could be used to reduce the power peak in fresh fuel elements. An extension of the computational methods to a mixture of two types of coated particles is being performed to look into possibilities for advanced fuel design.

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Table I: Some Data for Heterogeneous and Homogeneous C se, Changes During Burn-Up (signified by an arrow).

Case		I: UO_2	II: ThO_2-PuO_2	III: few Pu coat.part.
initial enrichment		8%	4%	4%
burnup(MWd/t)		o.o → 588oo.	o.o → 291oo.	o.o
k_{eff}	het	1.4o7o → 1.o417	1.11o → o.995	1.167
	hom	1.4o73 → 1.o415	1.1o2 → o.997	1.1o2
breeding Ratio	het	o.o → o.556	o.31o → o.756	o.192
	hom	o.o → o.557	o.321 → o.753	o.321
$\sigma_a(^{233}U)$	het	246.6 → 251.2	2o7.5 → 226.8	23o.1
	hom	246.5 → 251.2	21o.6 → 228.7	21o.6
$\sigma_a(^{235}U)$	het	2o4.9 → 217.9	149.3 → 2o2.8	194.o
	hom	2o6.1 → 218.9	15o.9 → 2o3.9	15o.9
$\sigma_a(^{239}Pu)$	het	126o.3 → 1268.3	8o2.7 → 1186.9	54o.8
	hom	1266.8 → 1275.1	816.6 → 1194.6	816.6
$\sigma_a(^{240}Pu)$	het	1o72.4 → 684.8	633.4 → 5oo.7	↑261.2
	hom	1o8o.8 → 7o7.2	667.o → 529.3	↑667.o
Pu composition at the end	het	43.o/25.2/19.6/12.3	12.o/31.2/29.5/27.4	
	hom	43.2/24.7/19.7/12.4	12.2/3o.3/3o.1/27.5	

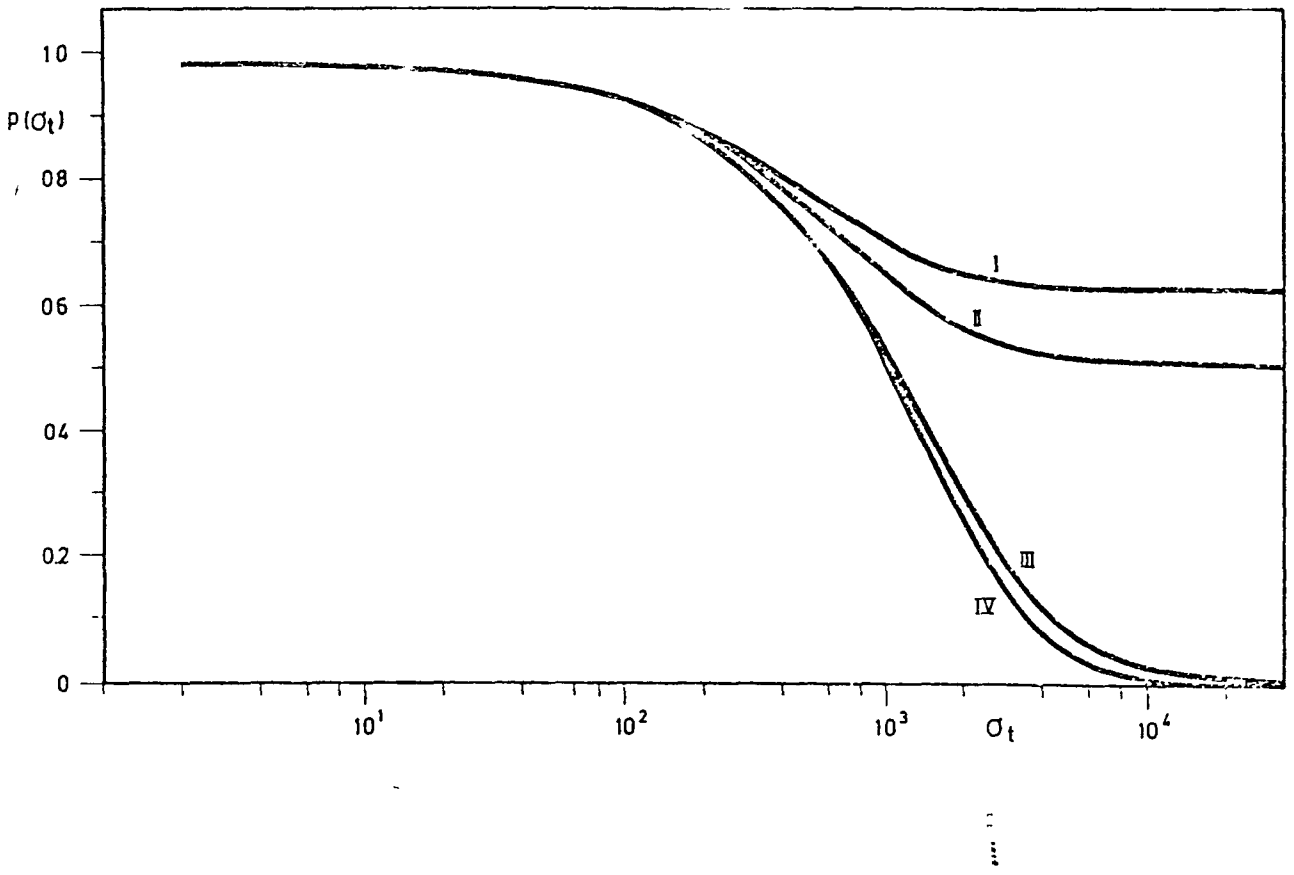

Figure 1: Penetration Probability as Function of Absorber Cross Section

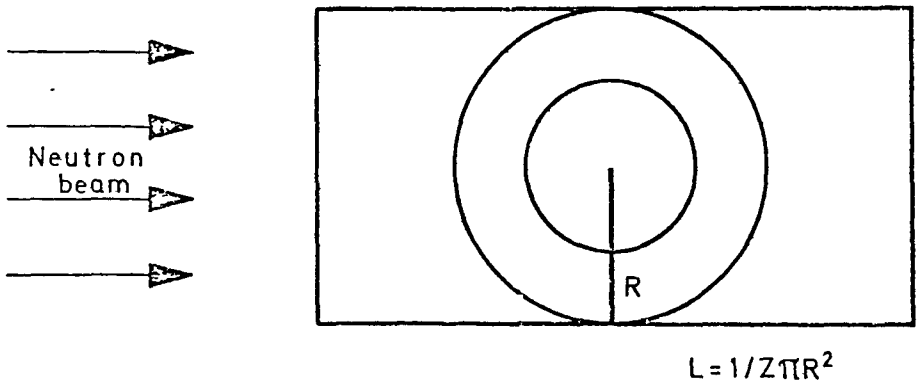

Figure 2: Model for a Neutron Beam through the Area of one Coated Particle

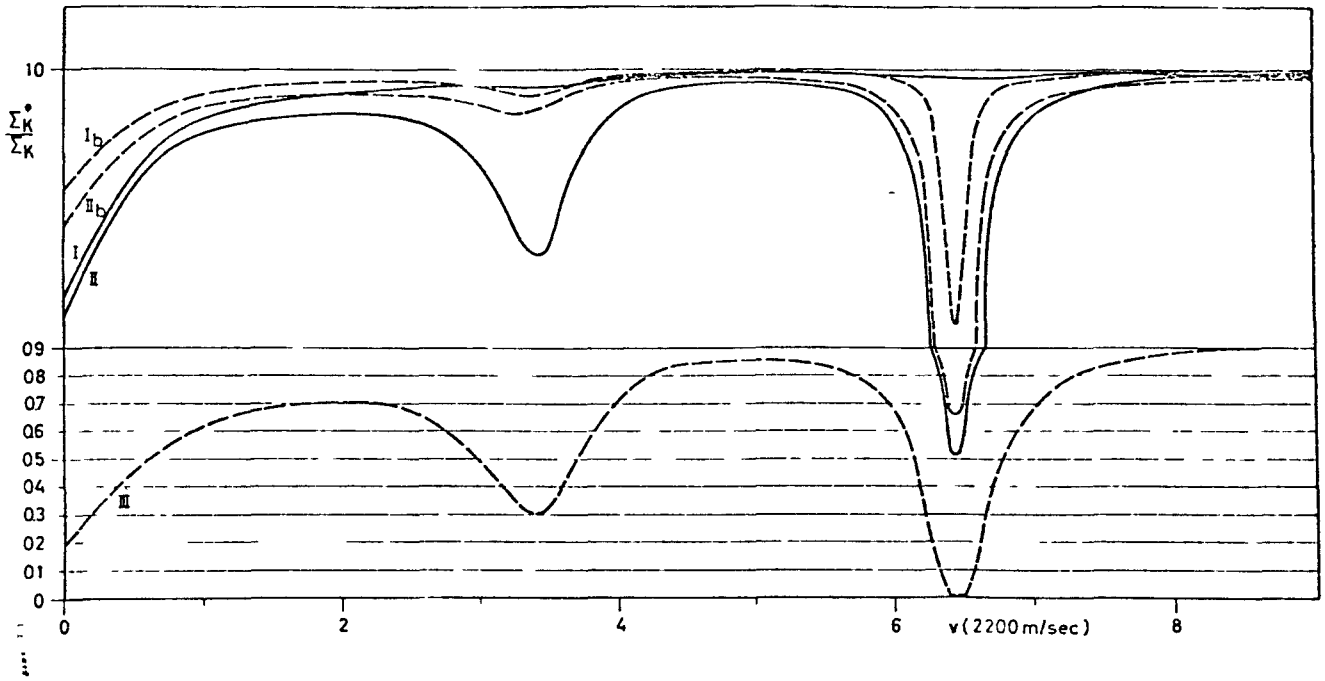

Figure 3: Ratio of the Effective Kernel Material Total Cross Section Σ_K^* to the Σ_K of Homogenized Medium.