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NEUTRON MULTIPLICATION AND SHIELDING PROBLEMS IN PWR SPENT-FUEL SHIPPING CASKS

C. DEVILLERS (C.E.A.-SACLAY)

P. BLUM (TRANSNUCLEAIRE)

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C. DEVILLERS - P. BLUM

1 - INTRODUCTION

To evaluate the degree of accuracy of computational methods used in the shield design of spent-fuel shipping casks, comparisons have been made between biological dose rate calculations and measurements at the surface of a cask carrying three PWR fuel assemblies.

Of importance for neutron shielding problems is the knowledge of (α, n) and spontaneous fission neutrons and of their multiplication in the subcritical fuel configuration ; an experimental method of control of multiplication factors has therefore been investigated. This method is based upon two series of measurements of the external neutron dose rate : the first measurements are made with the fuel carrying region filled with water, the second ones are made without water.

The shipping cask considered in this study is a TN-8 type cask, under normal transportation conditions, no water cooling is necessary to remove the decay heat. For safety reasons however, the neutron multiplication coefficient must remain below 0.95 in the unlikely event that the fuel assemblies are both non irradiated and plunged in water.

An experimental derivation of the keff in case of spent fuel assemblies plunged in water can therefore reveal the safety margin on the multiplication coefficient due to fuel depletion. In addition it can serve in checking the computational methods used in subcriticallity assessments.

2 - DESCRIPTION OF THE SHIPPING CASK

A cross-section of the TN.8 cask containing three PWR fuel assemblies is shown in Fig. 1. The 15 x 15 rod , 3.19 % enriched, fuel assemblies have been exposed to respectively 30760, 30560 and 29840 MWd/MTU during 953 days and cooled during a period of 243 days. The stainless steel walls of the fuel carrying cells are surrounded by boron carbide neutron absorbers. The biological shield consists of a lead region followed by a neutron shield composed of borated resin with copper fins inserted to transfer decay heat to the outside.

3 - *PRINCIPLE OF KEFF MEASUREMENTS*

The experiment consists of two series of neutron dose rate measurements performed at points A, B, C (and D symmetrical of C) indicated in Fig. 1. The two series differ between eachother by the presence or not of water in the fuel cells.

Let us call S_0 the spontaneous neutron source emitted by the spent fuel, k the multiplication coefficient, T the transmission coefficient from source to detector and M the measured neutron dose rate.

The transmission coefficient T is definied as the dose rate at a given measurement point resulting from a neutron source of one neutron per centimeter height per second in the three fuel assemblies. The measured neutron dose rate may therefore be expressed as :

$$M = \frac{S_0}{1 - k} T \qquad (1)$$

The ratio between two measurements at the same point with water and then without water can be written as :

$$\frac{M_1}{M_2} = \frac{1 - k_2}{1 - k_1} \cdot \frac{T_1}{T_2} \qquad (2)$$

where the subscripts 1 and 2 refer respectively to fuel with and without water.

The transmission ratio T1/T2 can be calculated accurately by neutron transport codes ; therefore equation (2) provides a relationship between k_1 (wet fuel) and k_2 (dry fuel).

If now the low value k_2 is known, even roughly, equation (2) gives an experimental value of k_1.

4 - CALCULATION OF TRANSMISSION COEFFICIENTS

The ratio of transmission coefficients depends essentially on the following features :

- attenuation of fast neutrons by water in case 1
- differences between spectra and spatial distributions of neutron sources due to different levels of induced fission neutrons in the two cases.

On the contrary, this ratio will not be affected by possible uncertainties in material compositions, neutron cross sections or detector response function.

The transmission coefficients have been calculated using the discrete ordinates transport code DOT-3 [1]. Adjoint calculations performed in the wet and dry cases provide a simple way of analyzing the sensitivity of transmission coefficients to energy and spatial distribution of the neutron source.

Figure 2. shows the geometrical treatment made in DOT calculations. Preliminary ANISN [2] calculations were done to condense the 100 groups DLC.2D library into a 23 energy group scheme. The neutron dosimeter is simulated by an adjoint point source at the right boundary of the cask, corresponding to the measurement point C :

$$S^{+}(G) = \frac{D(G)}{\Delta V(I_0, J_0)}$$

D (G) is the dosimeter response function and $\Delta V(I_0, J_0)$ the volume of spatial mesh containing the adjoint source.

The transmission coefficient is obtained by :

$$T = \frac{\sum_{I,J} S(I,J)\ \Delta V(I,J) \sum_{G} \phi^{+}(I,J,G)\, \chi(G)}{\sum_{I,J} S(I,J)\ \Delta V(I,J)}$$

where $\phi^{+}(I, J, G)$ is the adjoint flux, S (I, J) and $\chi(G)$ being the spatial and energy distributions of the neutron source in the fuel region.

Figures 3. and 4. illustrate the spatial dependence of the neutron importance with respect to dose in the case of wet and dry fuel assemblies :

$$I\ (I, J) = \sum_{G} \phi^{+}(I, J, G)\ \chi\ (G) \qquad (4)$$

These curves show in particular the much higher opacity to neutrons of the wet fuel.

Equation (3) has been used to study the sensitivity of the transmission coefficients to the spatial and energy distributions of the neutron source.

Neutron multiplication calculations which will be described later in this paper have been done in order to obtain realistic source distributions. It has been found , however, that the assumption of a uniform source distribution is quite acceptable as Table I shows.

TABLE I - SENSITIVITY OF TRANSMISSION COEFFICIENTS TO SPATIAL NEUTRON SOURCE DISTRIBUTION (235 U fission spectrum)

Spatial distribution	Transmission coefficient ($mremh^{-1}/n.cm^{-1}.s^{-1}$)		Ratio T1/T2
	T1 (wet)	T2 (dry)	
Realistic	$3.95\ 10^{-7}$	$1.142\ 10^{-6}$	0.346
Uniform	$3.98\ 10^{-7}$	$1.147\ 10^{-6}$	0.347

The sensitivity to energy spectra is illustrated in table II where have been considered the spectra of the different source terms arising either from spontaneous neutron emission (fission plus (α, n)) or from multiplication.

A realistic spectrum taking into account the contributions of these terms to the total source has also been derived from fuel depletion and multiplication calculations detailed in the following.

TABLE II - SENSITIVITY OF TRANSMISSION COEFFICIENTS TO NEUTRON SOURCE SPECTRA (uniform spatial distribution)

Spectrum	Transmission coefficient ($mremh^{-1}/n.cm^{-1}.s^{-1}$)		Ratio T1/T2
	T1 (wet)	T2 (dry)	
235U fission	$3.98\ 10^{-7}$	$1.15\ 10^{-6}$	0.347
239Pu fission	$4.19\ 10^{-7}$	$1.19\ 10^{-6}$	0.352
242Cm sp.fis.	$4.05\ 10^{-7}$	$1.16\ 10^{-6}$	0.349
244Cm sp.fis.	$4.12\ 10^{-7}$	$1.18\ 10^{-6}$	0.351
242Cm (α, n)	$2.46\ 10^{-7}$	$8.51\ 10^{-7}$	0.289
realistic	$4.04\ 10^{-7}$	$1.14\ 10^{-6}$	0.354

Exept for the (α, n) spectrum, there is not much difference among these results ; the transmission ratio corresponding to the realistic spectrum differs from the one obtained for Uranium 235 by less than 2 %. This conclusion should however be checked in case of lower burnup.

5 - DETERMINATION OF NEUTRON MULTIPLICATION COEFFICIENTS

5.1. *Fuel depletion*

The isotopic fuel composition after an irradiation of 953 days to an average burnup of 30400 MWd/MTU and a cooling time of 243 days has been calculated by the fuel depletion code APOLLO [3]. Results are reported in Table III.

TABLE III - COMPOSITION OF SPENT FUEL
(953 d irradiation, 30400 MWd/MTU, 243 d cooling)

Isotope	Weight (kg/MTU)
235 U	9.113
236 U	3.767
238 U	945.889
238 Pu	0.139
239 Pu	5.152
240 Pu	2.039
241 Pu	1.132
242 Pu	0.462
241 Am	0.066
243 Am	0.0065
242 Cm	0.0036
244 Cm	0.0123

5.2. *Calculation of neutron multiplication coefficients*

The DOT code has been used to determine theoretical multiplication coefficients of spent fuel assemblies in the shipping cask, with and without water.

In each case, two types of calculations have been performed : k-search and source plus multiplication. The second type is of course more realistic because the system is highly subcritical. Five group cross sections including two thermal groups were provided by APOLLO, first for fuel rod cells, and second for structural materials and neutron absorbers.

In the source plus multiplication calculation type, the spontaneous neutron source is taken to be uniform and k is defined as :

$$k = \frac{S_F}{S_0 + S_F}$$

where S_0 refers to the spontaneous and S_F to the multiplied neutron source.

Comparison of the results of the two different calculation types is reported in Table IV.

TABLE IV - MULTIPLICATION COEFFICIENT IN SHIPPING CASK (30400 MWd/MTU)

Calculation type	With water k_1	Without water k_2
k-search	0.6507	0.1589
source plus multiplication	0.6465	0.1589

These calculations provide almost identical results ; however the last ones will be kept for comparison with experimental values. It is the corresponding spatial source distribution that has been taken as the realistic distribution in Section 4.

Multiplication coefficients in fact vary in function of the location along the fuel assemblies due to spatial dependence of the fuel depletion. In order to compare with multiplication coefficients derived from measurements in the mid-region of the fuel assemblies it is necessary to correct the above results which correspond to the average burnup.

Correction factors to k in function of deviation from the average burnup have been calculated ; they are reported in Table V.

TABLE V - VARIATION OF k IN FUNCTION OF BURNUP

Burnup (MWd/MTU)	k/k (30500)	
	With water	Without water
27500	1.026	1.011
28500	1.017	1.007
29500	1.009	1.003
30500 (average)	1.	1.
31500	0.991	0.997
32500	0.983	0.993
33500	0.975	0.990

Sensitivities of k to burnup around 30500 MWd/MTU are 0.9 % per 1000 MWd/MTU with water and only 0.3 % per 1000 MWd/MTU without water.

5.3. *Measurement of multiplication coefficients*

Neutron dose rate measurements at points A, B, C (and D symmetrical of C) in the mid-plane of fuel assemblies have given the results of Table VI.

TABLE VI - NEUTRON DOSE RATE MEASUREMENTS

Location	Dose (count)		Ratio M2/M1
	With water (M1)	Without water (M2)	
A	2747	3392	1.23
B	2381	2881	1.21
$\frac{C + D}{2}$	2751	3469	1.26 ± 0.10

Dose rate measurements at the two opposite points C and D give an idea of the errors in measurement ratios M2/M1. This error is due to uncertainties in the exact location of the fuel assemblies inside their alveoles.

From equation (2) of section 3. one can write :

$$k_1 = \alpha k_2 + 1 - \alpha \qquad (5)$$

with $\alpha = \frac{T1}{T2} \frac{M2}{M1}$

When taking the calculated value of $k_2 = 0.157$, the transmission ratio calculated in section 4. : T1/T2 = 0.354 and the different measurement ratios of Table VI above, equation (5) gives the experimental values of k_1 shown in Table VII.

TABLE VII - EXPERIMENTAL VALUES OF THE MULTIPLICATION COEFFICIENT AT THE FUEL ASSEMBLY MID-PLANE (with water)

Location	k_1^{exp}
A	0.633
B	0.639
$\frac{C + D}{2}$	0.624 ± 0.030
average	0.628 ± 0.024

A theoretical evaluation of k_1 can be obtained from section 5.2. by taking into account the actual burnup of the mid-region of the fuel, about 10 % higher than the average one. The corresponding value is found to be $k_1 = 0.630$ which confirms the experimental derivation.

Similar comparisons between experimental and calculated multiplication coefficients in different locations along the fuel assemblies and for lower average burnups are being pursued.

Although the experimental value of k_1 in the fuel with water depends upon a calculation of k_2 in the fuel without water, it must be pointed out that the sensitivity of k_1 to an uncertainty in k_2 is low. In the experiment considered here :

$$\frac{\Delta k_1^{exp} / k_1^{exp}}{\Delta k_2^{cal} / k_2^{cal}} = 0.11$$

that is to say an error of 10 % in the calculated k_2 would result in an error of 1 % in the experimental k_1 which is below the other sources of uncertainty.

We have therefore demonstrated by this example that the proposed experimental method of control of multiplication coefficients in shipping casks is feasible and provides keff values in good agreement with theoretical calculations. Because one series of measurements is made without water in the fuel assemblies this method applies only to shipping casks that can remove decay heat without water cooling.

6 - *SHIELDING CALCULATIONS*

Now that the neutron multiplication is proved to be correct, neutron dose rate measurements can be used to check the calculations of spontaneous neutron sources and transmission coefficients which are both necessary in designing spent fuel shipping cask shields. Additional gamma dose rate measurements will also allow us to verify the accuracy of gamma sources and gamma transmission calculations.

6.1. *Prediction of neutron dose rates*

The spontaneous neutron source for the average burnup of 30400 MWd/MTU has been evaluated from the average isotopic composition of the spent fuel (Table III). The different source terms are shown in Table VIII together with the nuclear data utilized.

TABLE VIII - SPONTANEOUS NEUTRON SOURCE (30400 MWd/MTU, 243 d COOLING)

Isotope	Half-life (y)		Neutron yield		Neutron source ($n.g_U^{-1}.s^{-1}$)	
	α	s.f.	10^{-8} n/α	ν	(α, n)	s.f.
238 Pu	87.75	5.10^{10}	2.52	2.26	2.2	0.4
239 Pu	$2.44\ 10^{4}$	$5.5.\ 10^{15}$	1.99	2.15	0.2	-
240 Pu	$6.54\ 10^{3}$	$1.32\ 10^{11}$	1.99	2.17	0.3	1.8
242 Pu	$3.87\ 10^{5}$	7.10^{10}	-	2.20	-	5.7
241 Am	$4.32\ 10^{2}$	$2.3\ 10^{14}$	2.52	2.43	0.2	-
242 Cm	0.446	$6.6\ 10^{6}$	3.47	2.57	15.5	77.6
244 Cm	17.8	$1.346\ 10^{7}$	3.02	2.75	1.1	136.0
					19.5	221.5

The (α, n) yields in oxide are obtained from yields in pure oxygen[4] multiplied by the fractional stopping power of oxygen in uranium oxide with respect to alpha particles.

Neutrons from (α, n) reactions on ^{18}O contribute only 8 % to the total emission whereas the most important terms come from spontaneous fissions of ^{242}Cm (32 %) and ^{244}Cm (57 %). The total spontaneous neutron source is equal to 240 neutrons per gram uranium per second.

In case of normal transportation conditions, without water in the fuel carrying region, the neutron source per unit length is therefore :

$$S = S_0 + S_F = 9.83\ 10^{5}\ n.cm^{-1}.s^{-1}$$

This value has to be corrected for non-uniform burnup effects in order to compare dose predictions with dose measurements in the mid-plane of the cask. Table IX provides correction factors in function of deviation from the average burnup.

TABLE IX - VARIATION OF TOTAL NEUTRON SOURCE VERSUS BURNUP (fuel without water, 243 d cooling)

Burnup	S/S (30500)
27500	0.71
28500	0.80
29500	0.90
30500 (average)	1.
31500	1.11
32500	1.23
33500	1.36

The sensitivity of the neutron source to burnup around 30500 MWd/MTU is 12 % per 1000 MWd/MTU.

Because we are interested in an absolute comparison, two additional corrections must be done :

a/ correction of transmission coefficient due to few group cross sections used in DOT 3 calculations : comparison between 100 groups and 23 groups ANISN calculations in cylindrical geometry gives a correction factor of 1.22 at the external surface of the cask.

b/ corrections of measurements due to the dosimeter size (20.8 cm diameter) to convert the experimental values at 10.4 cm from the cask surface into corresponding values at the surface ; these corrections have been estimated by MERCURE 4[5] in using a dose point kernel fitting ANISN calculations. Correction factors were found as 1.13, 1.14 and 1.21 respectively for points A, B, C, (D).

Neutron dose rate predictions in the mid-plane of the cask with a neutron source corresponding to 33500 MWd/MTU (burnup of the mid-region of the fuel assemblies), are compared with corrected experimental values in Table X.

TABLE X - COMPARISON OF NEUTRON DOSE RATE PREDICTIONS WITH EXPERIMENT (mid-plane, without water)

Location	Dose rate ($mremh^{-1}$)	
	calculation	experiment
A	1.98	1.81
B	1.37	1.60
$\frac{C + D}{2}$	1.88	2.00

From this comparison it can be concluded that calculational methods used in neutron shielding problems are sufficiently accurate.

6.2. *Prediction of gamma dose rates*

Gamma sources due to fission product decay in the mid-region of the fuel assemblies have been calculated by the computer code PEPIN 76[6] and are reported in Table XI.

TABLE XI - GAMMA RAY SOURCES IN SPENT FUEL (33500 MWd/MTU, 243 d COOLING)

Energy (MeV)	$Gamma.g_U^{-1}.s^{-1}$
2.75 - 2.25	$4.43\ 10^6$
2.25 - 1.75	$2.34\ 10^8$
1.75 - 1.25	$4.46\ 10^8$
1.25 - 0.75	$1.29\ 10^{10}$
0.75 - 0.507	$1.55\ 10^{10}$

Gamma dose rates were calculated at points A, B, C by MERCURE 4[5] in the exact geometry of the shipping cask. Table XII shows the comparison between the results obtained and the experimental values.

TABLE XII - COMPARISON OF GAMMA DOSE RATE PREDICTIONS WITH EXPERIMENT (mid-plane, without water)

Location	Dose rate ($mremh^{-1}$)	
	Calculation	Experiment
A	21.6	20.
B	20.5	28.
$\frac{C + D}{2}$	25.0	28.

As for neutrons the accuracy of theoretical predictions is shown to be sufficient. A reason for the slight underestimation of calculations could be the homogeneous treatment of cooling copper fins around the cask.

7 - CONCLUSIONS

7.1. The proposed experimental method consisting of measuring neutron dose rates outside a PWR spent fuel shipping cask, fuel being successively wet and dry, permits to obtain a measure of the keff of wet fuel assemblies

7.2. In case of a TN-8 type container carrying three subassemblies having an average burnup of 30400 MWd/MTU the experimental method provides keff with an accuracy of 0.024 ; the uncertainty is essentially due to lack of knowledge about the exact location of the fuel assemblies in their alveoles.

7.3. The experimental derivation of keff corresponds to that region of the fuel which is seen by the neutron dosimeter. Measurements at different locations along the fuel assemblies can be processed by the same method. The analysis of measurements is easier if the burnups of the different fuel assemblies are close together.

7.4. Neutron multiplication coefficients provided by the APOLLO and DOT 3 codes are located within the uncertainty range of the experimentally derived values.

7.5. Neutron transmission coefficients involved in the experimental method and in neutron dose rate predictions are little sensitive to spatial and energy distributions of neutron sources and can be calculated once and for all.

7.6. The computer codes APOLLO plus DOT for neutron source calculations and ANISN plus DOT for neutron transmission calculations provide neutron dose rate predictions in agreement with measurements to within 10 %.

7.7. The PEPIN 76 code used in deriving fission product decay gammas and the point kernel code MERCURE 4 treating the gamma-ray transmission give gamma dose rate predictions , which generally differ from the measurements by less than 25 %.

REFERENCES

1. F.R. MYNATT, W.W. ENGLE, M.L. GRITZNER, W.A. RHOADES, and R.J. RODGERS "The DOT III Two-Dimensional Discrete Ordinates Transport Code", ORNL-TM-4280 (1973)

2. C. DEVILLERS "Système ANISN - Description et mode d'utilisation du programme aux ordonnées discrètes ANISN et des programmes auxiliaires", Note CEA.N.1358 (1970)

3. A. HOFFMANN, F. JEANPIERRE, A. KAVENOKY, M. LIVOLANT, H. LORAIN, "APOLLO - Code multigroupe de résolution de l'équation du transport pour les neutrons thermiques et rapides", Note CEA.N.1610 (1972)

4. E.M. TZENTER, A.B. SILIN, "Rendement de la réaction (α, n) en fonction de l'énergie des particules α ", Atomnaya Energia vol.19 n°1 (1965)

5. C. DEVILLERS, C. DUPONT, "MERCURE 4, Un programme de Monte Carlo à trois dimensions pour l'intégration de noyaux ponctuels d'atténuation en ligne droite", Note CEA.N.1726 (1974)

6. To be published

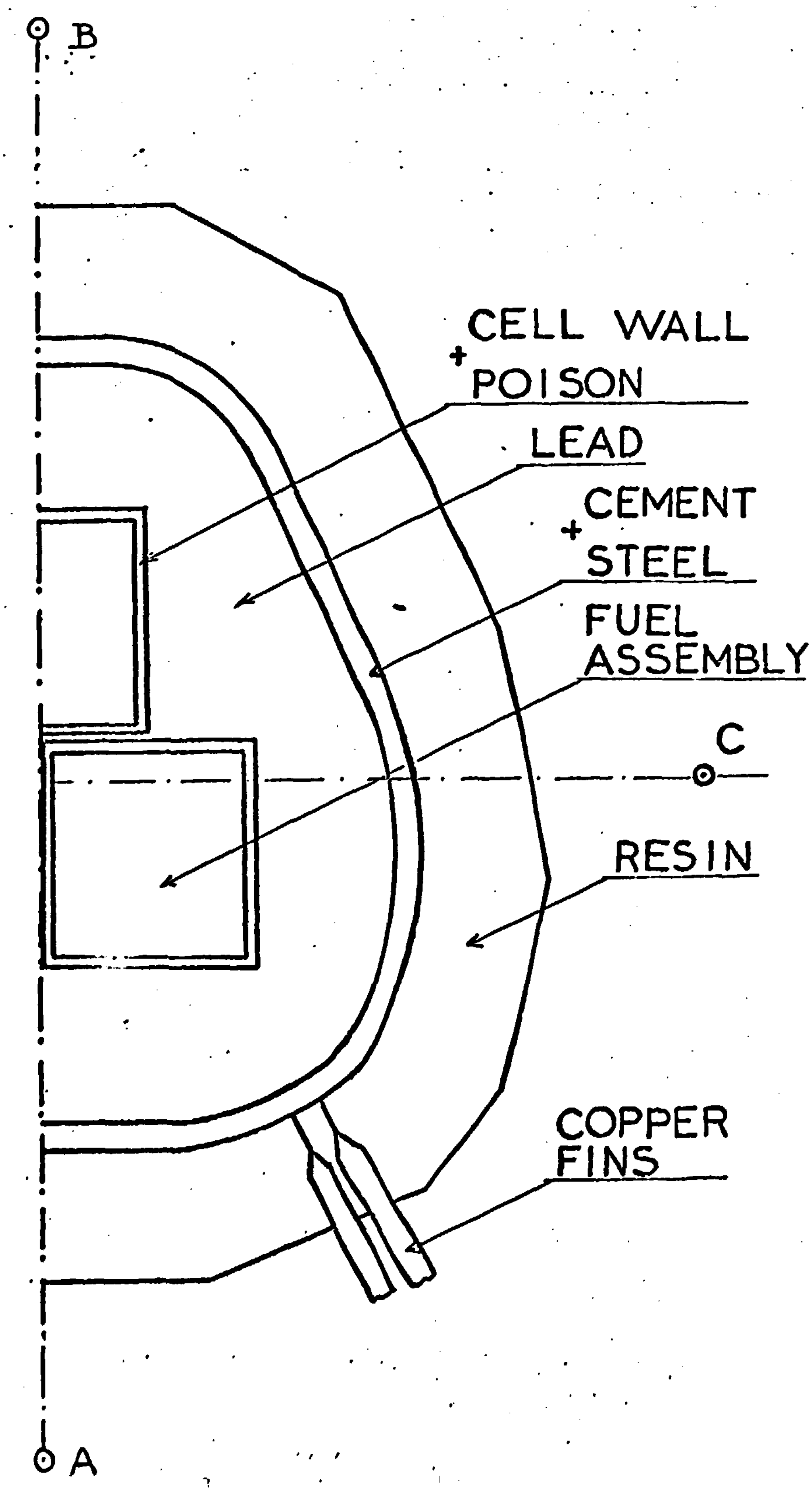

Fig.1 CROSS SECTION OF TN-8 SHIPPING CASK

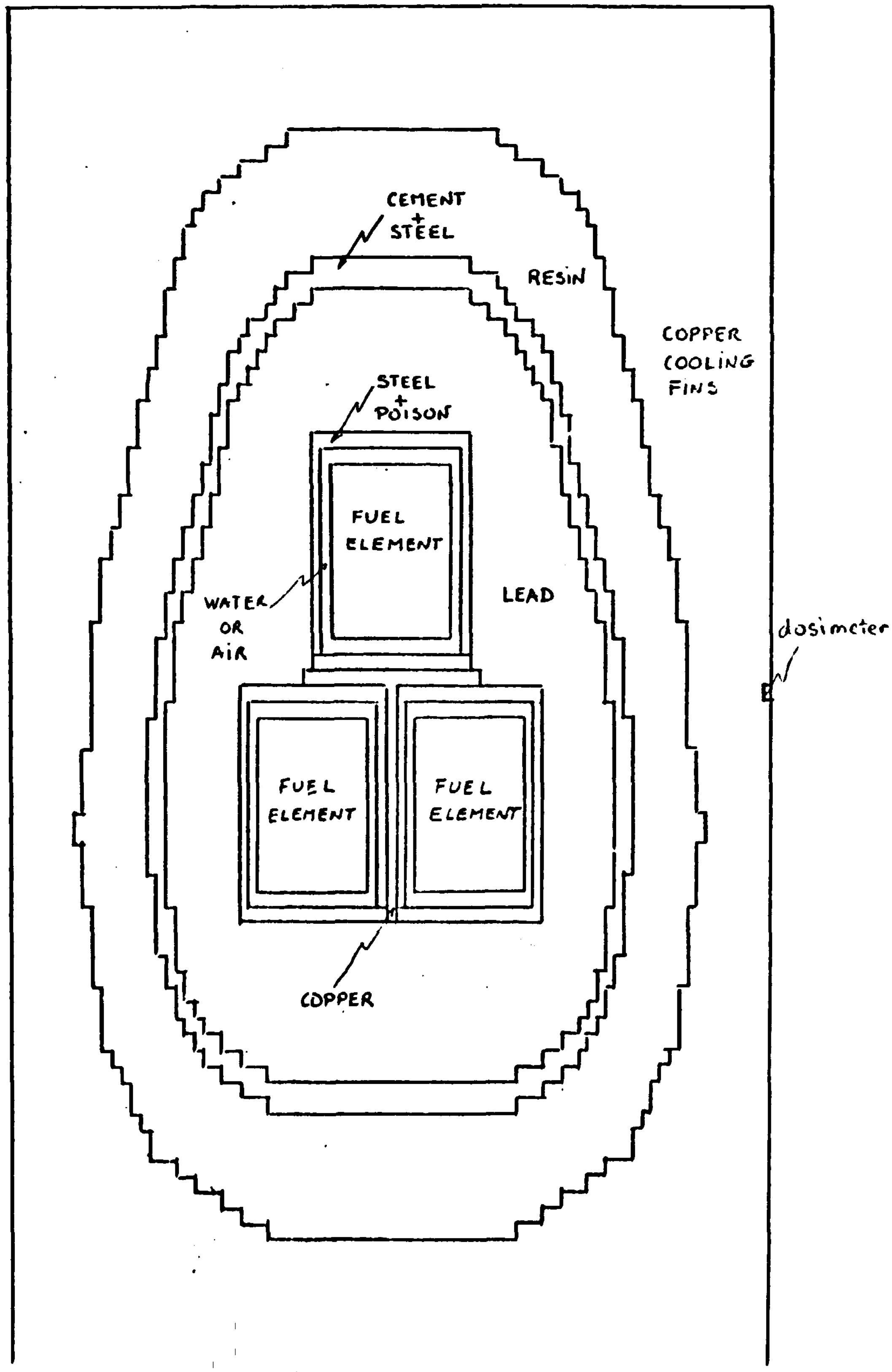

Fig.2 GEOMETRY OF DOT III TRANSMISSION CALCULATIONS

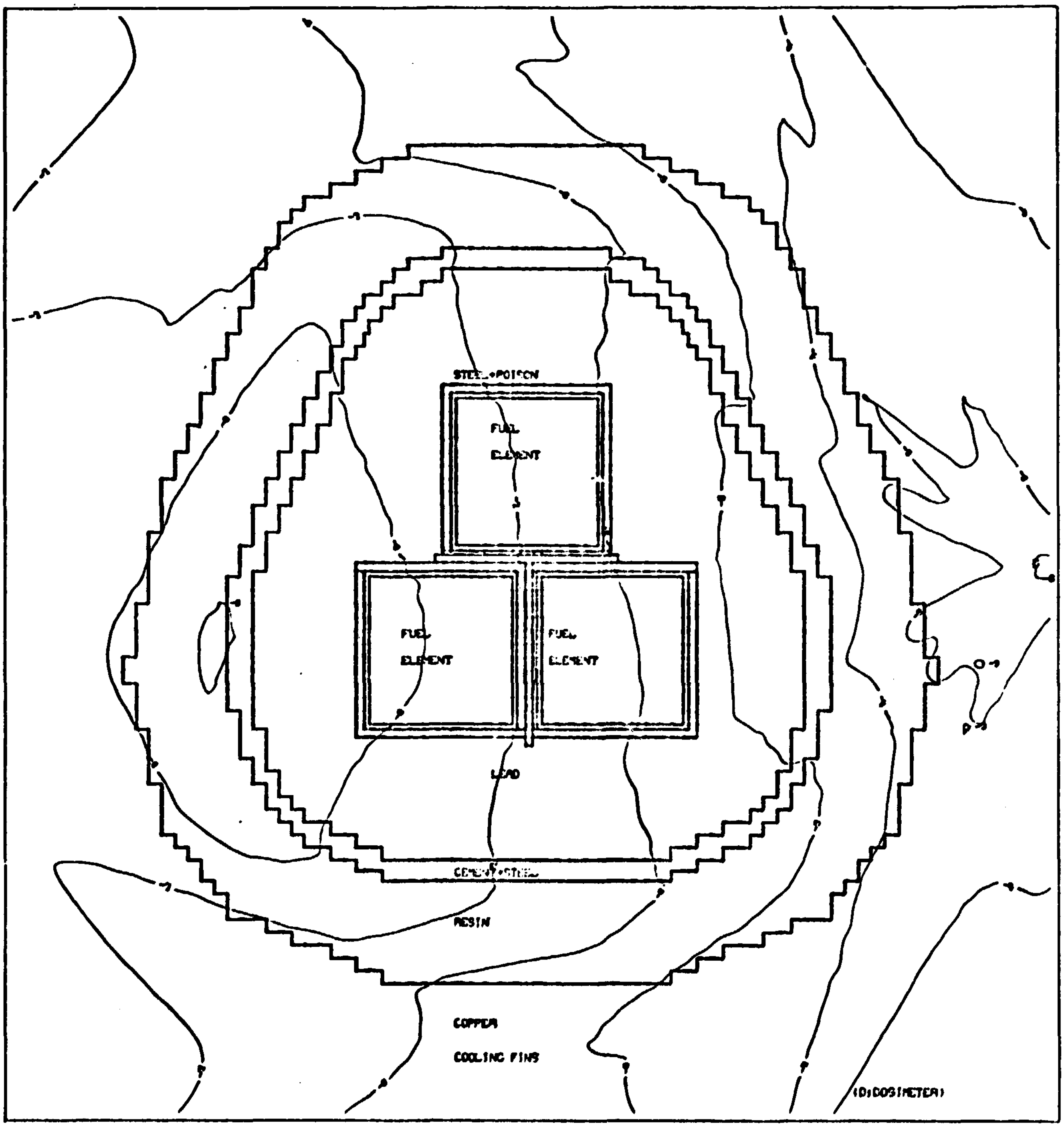

Fig.3 IMPORTANCE OF NEUTRONS VERSUS SPACE WITH RESPECT TO DOSE

FUEL WITH WATER

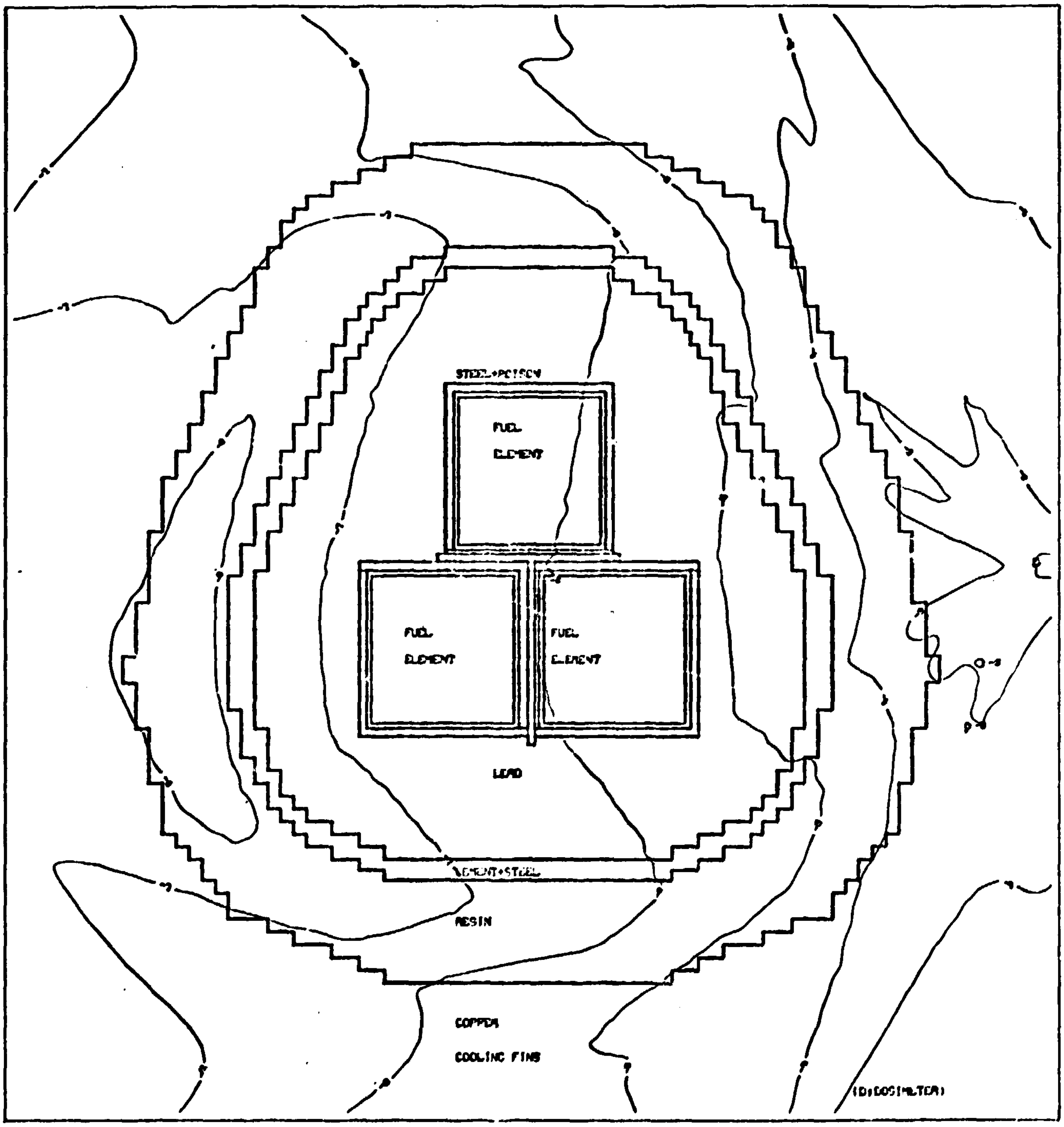

Fig.4 IMPORTANCE OF NEUTRONS VERSUS SPACE WITH RESPECT TO DOSE

FUEL WITHOUT WATER