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Studies of the Effect of Heavy Water in the Fast Reactor FRO

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STUDIES OF THE EFFECT OF HEAVY WATER IN THE FAST REACTOR FR0

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ABSTRACT

Core 9 of the FR0 fast critical assembly was diluted with heavy water to 24 vol. per cent, contained in thinwalled copper cans. The report describes measurements of the critical mass and the reactivity coefficient of heavy water in this core. The effect of the heterogeneous core composition on these items is also dealt with. The results are compared with theoretical predictions using several computer codes Criticality is accurately predicted, but the measured reactivity coefficient of heavy water is about 20 % lower than the value obtained with the best available methods, involving the SPENG and DTF-4 programmes.

The result of bunching measurements, in which the degree of heterogeneity of core composition was changed, is compared with theoretical estimates of the resonance shielding, flux advantage and leakage components of the heterogeneity effect.

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CONTENTS

1. INTRODUCTION

Current studies of steam cooling for fast reactors in Sweden include heavy water steam as an alternative coolant [1]. The interest in heavy water in this context stems from the fact that the neutron energy spectrum would be harder than with light water steam, and this would result in a higher breeding ratio. Predictions indicate a lower loss-of-coolant reactivity with heavy water steam which also would be favourable [2, 3]. In order to support these studies the effect of heavy water was determined experimentally in core 9 of the FR0 fast critical assembly. It was diluted with heavy water to 24 vol. per cent. The experiments, together with results and some comparisons with theoretical predictions, are described in the present report. Most of the experiments were concerned with the D_2O-void reactivity and were made in order to check the accuracy of theoretical predictions. This study is a complement to similar work on the reactivity of polythene (CH_2) in FR0 cores 5 and 7 described in AE-305 [4], to which we shall frequently refer.

In current work on fast critical assemblies the intermediate energy neutron region ($\sim$ 0.1 - 100 keV) is receiving increased attention. In order to obtain soft neutron spectra, in which this region is emphasized, graphite, beryllium and polythene have been used as diluents. Heavy water is here an alternative with some advantages. It is a much more effective moderator than graphite. It is less effective than polythene or other hydrogen-containing compounds, but the complications which accompany hydrogen, viz. anisotropic scattering and thermalization, are less significant. Deuterated polythene or other organic compounds have been contemplated as an alternative, but their fabrication is very expensive. The effects of graphite, heavy water and polythene on the critical mass and spectrum of various FR0 assemblies are shown qualitatively in Figure 1. The arrows show that large spectrum degradations can be achieved with polythene and heavy water without exceeding the fuel inventory. With graphite, on the other hand, only a minor softening of the spectrum is possible. It should be pointed out that the relations shown in Figure 1 are to some extent peculiar

to the reactor studied and depend on the magnitude of the leakage component and the composition of the system.

A description of the FR0 reactor is given elsewhere [5].

2. DESCRIPTION OF HEAVY WATER DILUTED ASSEMBLY AND DETERMINATION OF CRITICAL MASS

The core of the D_2O-diluted FR0 assembly 9 was cylindrical with core height = 34.4 cm and equivalent (smoothed) diameter = 40.4 cm. It was surrounded by the usual thick (~ 35 cm) copper reflector. The heavy water was contained in thin-walled (0.2 mm) copper cans with dimensions 0.355 x 4.3 x 34.4 cm^3 extending through the core. The core was built up in the pattern shown in Figure 2, 0.71 cm thick layers of uranium fuel (20 % U235) alternating with 0.315 cm thick layers of heavy water (excluding the thin copper cans and steel frames of the fuel elements).

The composition of core 9 is given in Table 1.

Table 1: Composition of FR0 core 9

Material	Vol. %	Atomic density $10^{22}/cm^3$
Uranium	58.5	
U235		0.568
U238		2.234
Steel	6.5	
Fe		0.408
Cr		0.096
Ni		0.048
D_2O-cans	29.2	
D		1.595
O		0.798
Cu		0.303
Void	5.8	

The loaded uranium mass was corrected for imperfections in the geometry including the core edge irregularity and the mass equivalence of the excess reactivity. The critical mass of the heterogeneous core determined in this way is 96.0 kg U235. In order to obtain a value which can be compared with calculations for a homogeneous model a correction was also made for the heterogeneity of the core. The correction was obtained by "bunching" measurements in which the thicknesses of the fuel and heavy water layers were increased in a quadrant of the core or in the whole core. The reactivity changes due to changes in layer thickness were determined by means of calibrated control rods. The results are shown in Figure 3, in which the reactivity of the assembly is given as a function of fuel layer thickness. By linear extrapolation to zero thickness the reactivity loss compared with the normal arrangement is estimated as 0.22 ± 0.05 %. The error margin includes the estimated uncertainty due to the slight change in the distribution of the fuel which accompanies bunching. The reactivity worth of core material relative to reflector material at the core-reflector interface was found to be 0.20 ± 0.02 % per kg U235. These data give a heterogeneity correction to the critical mass of 1.1 ± 0.3 kg U235. The critical mass of the equivalent homogeneous core is then 97.1 ± 0.5 kg U235. The quoted error arises from the heterogeneity correction and the uncertainty in the enrichment of the FR0 fuel (cf [6]).

A comparison with the theoretical predictions of criticality and heterogeneity effect is postponed to Section 5.

3. MEASUREMENTS OF THE D_2O-VOID REACTIVITY EFFECT

Method of measurement

The reactivity worth of heavy water versus void was determined by replacing one or several D_2O-cans by similar cans which were empty. Reactivity was measured by the doubling time technique in a manner described elsewhere [7]. When converting doubling time to reactivity an effective β-value of 0.78×10^{-2} was used, which is consistent with a calculation on core 9 giving $\beta_{eff} = 0.785 \times 10^{-2}$. The uncertainty in β will not be included in the errors pertaining to the experimental results.

The experimental method was similar to that used in the polythene-void measurements [4]. As in those experiments the observed fluctuations in reactivity were consistent with a standard error in a measured reactivity difference of not more than $\pm 1 \times 10^{-5}$. This corresponds to a relative error of less than ± 3 % in most of the experimental results given here. Other sources of error in these experiments are insignificant. The heavy water contents of the cans were accurately known (mean value: 47.6 ± 0.5 g D_2O) and recorded. The light water content was checked by infrared analysis and was found to be less than 0.1 % [8]. The effect of this slight impurity can be ignored. The largest difference in copper weight between two cans was 1 gram. Since the reactivity worth of copper is less than 10^{-6} per gram [9] this difference is negligible.

The errors assigned to the experimental results below are the estimated standard errors.

Experimental results

The effect of heavy water was measured with D_2O-cans located in the core at different radial distances from the centre. The integrated effect over the whole core height was obtained in this way; measurements were also made in the axial reflector above and below the core. In a separate experiment D_2O-cans were replaced by empty cans uniformly over a quadrant of the core in order to determine the average reactivity coefficient in the core.

Heterogeneity effects were estimated in supplementary measurements in which two or four D_2O-cans were bunched together. The result of this procedure applied to the central fuel element is shown in Figure 4. It is given in terms of the reactivity coefficient of heavy water (10^{-5}/g D_2O). The increase in the coefficient with increasing thickness of the heavy water layer is attributed to several phenomena. The fast fission rate is enhanced in the surrounding bunched fuel plates and the resonance capture in U238 is reduced. Neutron streaming in the empty cans also contributes, the negative reactivity effect of streaming being largest in the widest channel. Streaming effects

have been measured in the first FR0 core using channels of similar shape [10]. Results from those measurements indicate that not more than about 50 % of the heterogeneity effect in the present experiment can be attributed to neutron streaming.

The reactivity coefficient valid for a uniform (homogeneous) injection of heavy water in the central fuel element is obtained by linear extrapolation of the results in Figure 4 to zero thickness of the heavy water layer. The value obtained is $(0.765 \pm 0.015) \times 10^{-5}$/g, which is 3.7 % lower than the value measured with the single D_2O-can.

Bunching measurements were also made in the end and side reflectors. The reactivity coefficient proved to be very small in the reflector, however, and so the heterogeneity effect could not be resolved.

The results from measurements at various distances from the core centre are summarized in Figure 5. The experimental values were obtained with single D_2O-cans in the different positions and were reduced by 3.7 % to correct for heterogeneity. The spatial dependence of the reactivity coefficient is similar to that of polythene in FR0 assemblies 5 and 7 with one notable exception. While the effect of heavy water versus void is positive in all parts of the assembly the polythene-void effect becomes negative in the reflector. The positive effect arises from the collision of neutrons with the added heavy water (or polythene) and subsequent scattering back into the core. In the case of collisions in hydrogen, however, the large energy loss increases the probability of capture in the copper reflector, and this effect dominates in the case of polythene.

The results shown in Figure 5 are valid for D_2O-slabs extending through the core only. Measurements in the end reflector show that the reactivity coefficient of a slab extending right through the fuel element, including top and bottom reflectors, is (10 ± 2) % higher.

The average reactivity coefficient in the core was determined by replacing one D_2O-can in each fuel element in an approximate quadrant of the core (except the central fuel element, cf Figure 2)

by an empty can. The D_2O-content was thereby reduced by 25 % in the quadrant. A value valid for the whole core was determined from the measured reactivity change, assuming the contributions from all quadrants and the central fuel element to be additive. The result can be compared with the value obtained by numerical integration over the reactor core of the curve given in Figure 5. The following numbers were obtained:

Arrangement	Reactivity coefficient 10^{-5}/gram D_2O
Measurements in a quadrant of the core	0.527 ± 0.005
Measurements with single D_2O-cans	0.51 ± 0.01

The errors here do not include the uncertainty due to the irregularity of the core boundary. The result from the measurements on single cans involved an error in the numerical integration which is neither included in the stated error. These items may contribute uncertainties of the order of (1-2) % in each of the values. The agreement between the results confirms that the effects of small amounts of heavy water are additive.

4. THEORETICAL METHODS AND DATA

The theoretical work is based on the programme SPENG and its related data library [5]. The latter has been revised on several occasions during recent years. The data underlying this study are given in Refs. 5, 11, 12 and 13 . The conclusions to be drawn from comparisons with the experimental results are qualitative rather than quantitative and are not focused on some particular cross section. Therefore no detailed specification of the cross sections is given here. The data are the same as those used in the study of the polythene-void reactivity effect [4]. This should make it possible to compare the results of the studies of heavy and light hydrogen in the FR0 reactor.

Cross sections for deuterium have not been much tested in fast reactor work and therefore deserve particular attention in this study. Scrutiny of the SPENG library shows that the elastic scattering and (n, 2n) cross sections given for deuterium are in good agreement with BNL 325 of 1958 [14] and 1964 [15], respectively. The first Legendre moments (centre of mass system), which appear in the transport cross section, are in fair agreement with differential data given in BNL 400 of 1962 [16] (cf Section 5).

In the SPENG programme the neutron spectrum of the homogenized bare core is calculated at typically 300-500 energy points using the B1 approximation of the Boltzmann equation. The spectrum obtained from SPENG for FR0 core 9 is shown in Figure 6. Multigroup cross sections and other data for use in subsequent calculations on the reflected assembly or on the heterogeneous plate lattice are also computed. In the present study the number of energy groups was 16.

Data from SPENG are transferred by tape or punched cards into the multigroup programme MONDAY (diffusion theory, [17]) or DTF-4 (S_4 approximation, [18]) for reactivity calculations on the reflected assembly. These programmes are one-dimensional. They were mostly run for cylindrical geometry, the axial leakage being accounted for by a buckling parameter B_z^2.

An attempt was made to calculate the effect of heterogeneity on reactivity (critical mass). The effect can be divided into three parts - the resonance component, the flux advantage component and the leakage component. The resonance component - the difference in resonance shielding in the homogeneous and heterogeneous cores - can be estimated by means of a heterogeneity routine in SPENG. The isotopes concerned are U235 and U238. Use of equivalence theory reduces the problem to the determination of an equivalent background scattering cross section. A formula given by Hummel et al. [19] is employed at this point. It allows for the mutual shielding between the fuel plates.

The flux advantage component is defined here as the reactivity effect of the broad-energy-group flux variations in the plate lattice. In particular, according to experimental information [20], the flux in the MeV region is higher in the fuel than in the heavy water layers, and this leads to an increased U238 fast fission rate as compared with conditions in the homogeneous medium. The flux advantage component is calculated with the FLIS programme, which computes fluxes and reactivity in a repetive slab lattice in up to 22 energy groups and 10 different materials. FLIS is an integral transport theory programme using the collision probability method. It is based on the work by I. Carlvik [21] and was coded by Å. Ahlin.

FLIS has recently been modified in such a way as to permit the leakage parallel to the slabs to be calculated. The spatial flux distribution in this direction is assumed to be given by the Bessel function, $J_o(\beta r)$; the leakage is thus determined by the buckling β^2. It can be expressed by the formula

$$\text{Leakage} = D_{11}\,\beta^2\,\phi$$

where the symbols have their usual meaning. The leakage parallel to the layers in the FR0 core configuration (Figure 2) can similarly be written

$$D_{11}\,\frac{1}{2}\,B_r^{\,2} + D_{11}\,B_z^{\,2},$$

since one half of the radial leakage occurs in the direction parallel to the layers (this formula is strictly valid only in the homogeneous case).

Putting the two expressions equal, one obtains

$$\beta^2 = \frac{1}{2}\,B_r^{\,2} + B_z^{\,2}$$

The bucklings $B_r^{\,2}$ and $B_z^{\,2}$ were estimated from the physical dimensions of the core and an assumed value of the reflector saving (cf. below). Since the various layers were about one neutron mean

free path in thickness, or thinner, variations in leakage perpendicular to the layers were ignored.

The D_2O-void reactivity effect was studied by full criticality calculations using the MONDAY programme. Complementary runs were also made with DTF-4. Although a perturbation method could alternatively have been used, as the considered reactivity changes were small, the criticality programme MONDAY was preferred because a) it gives in principle a more accurate result and b) it can be linked with SPENG and is very fast. A perturbation code based on transport theory was not available.

All programmes described here were written for the IBM 7044 computer. Further details and comments concerning the theoretical methods will be given in the following section.

5. COMPARISON OF EXPERIMENT AND THEORY

Criticality including the heterogeneity effect

The diameter of the equivalent homogeneous core 9 with a smooth boundary was determined experimentally as 40.4 cm (cf Section 2). The results of the criticality calculations with 16 energy groups on this system are listed in Table 2 .

Table 2: Result of criticality calculations

Code	Geometry	k_{eff}
MONDAY	Cylinder Sphere	0.999 1.001
DTF-4	Cylinder Sphere	1.007 1.009

In the calculations using cylindrical geometry the axial buckling B_z^2 was computed with the reflector saving taken as 11.0 cm. This value is the average for the copper reflector obtained by meas-

urements in FR0 assemblies with different neutron spectra [22]. When spherical geometry was considered the core radius was determined by applying a shape factor for critical mass according to Davey [23].

The k_{eff} values are close to 1, but this cannot be interpreted as an indication of an overall accuracy of the cross sections used. They form a consistent set, however, which yields k_{eff} values near 1 for most FR0 assemblies. The S_4 calculation (DTF-4) predicts a more reactive system (0.8 % Δk), which appears to be a systematic trend in connection with FR0 assemblies. The outputs from the computer show that the higher k_{eff} value of DTF-4 is due to a lower leakage, but there is no appreciable difference in the neutron spectra computed by the two programmes.

Next, the predicted effect of heterogeneity on reactivity will be examined. The results of SPENG-FLIS calculations are presented in Table 3 and plotted in Figure 3 together with the experimental results.

Table 3: Effects of heterogeneity on reactivity

Calculated effect	Method	Reactivity change, 10^{-5} Fuel plate: 0.71 cm → 2.84 cm
Resonance, U235	SPENG	- 41
Resonance, U238	SPENG	215
Flux advantage	FLIS	908
Leakage	FLIS	-572
Total	SPENG-FLIS	510
Total	Experiment	668 ± 40

The numbers listed in the table are the reactivity increments obtained when bunching the fuel and heavy water layers (uranium = = 0.71 cm, D_2O = 0.315 cm) into layers of four times the normal thicknesses. In order to reduce the FLIS calculation to a three-region problem the thin copper cans and steel frames, which together comprise 9.8 % of the core volume, were treated as a single homogenous region. In the SPENG calculation of resonance shielding the use of a rational approximation for the escape probability introduces a source of error. These items may account for the difference between the experimental and theoretical results. It may be noted that the calculated effect is split into components which are of equal order of magnitude but have opposite signs.

The scattering cross sections used in FLIS are in general reduced by a transport correction, i.e.

$$\Sigma_{tr} = \Sigma_{tot} - \bar{\mu} \Sigma_s$$

A separate run, in which isotropic cross sections were used for heavy water ($\bar{\mu} = 0$), reduced the leakage component by only 28×10^{-5}. This shows that the theoretical result is fairly insensitive to the manner of accounting for anisotropic scattering.

The D_2O-void reactivity effect

The average reactivity coefficient of heavy water in the core was calculated by SPENG-MONDAY or SPENG-DTF-4 with the normal heavy water density in the core and with this density reduced by 20 %. The effect of the density change on the spectrum is illustrated in Figure 6.

The radial distribution of the reactivity effect was calculated by removing the heavy water content in annular zones 1 cm wide, located at various distances from the core centre. The results, expressed in terms of the reactivity coefficient, are plotted together with the experimental values for the D_2O-cans in Figure 5. Since the measurements were corrected for the finite thickness of the

D_2O-can, and since the effects of small amounts of heavy water were additive, the results in Figure 5 should be directly comparable. A theoretical check on additivity was obtained by comparing the integrated effect of the removal of heavy water in annular zones with that of the uniform density change. Values of the average reactivity coefficient are compared in Table 4. The first two MONDAY calculations confirm the assumption of additivity.

Table 4: Reactivity coefficients of heavy water
Comparison of results

Item		Reactivity coefficient 10^{-5}/gram D_2O
MONDAY calculations	Uniform density change	0.667
	Annular zones	0.660
DTF-4 calculation	Uniform density change	0.641
Experiment		0.52 ± 0.01

All calculations predict a larger reactivity coefficient than the experimental value.*) To discuss possible causes for the discrepancy the effect of heavy water (versus void) is divided into four components:

1) Decreased leakage due to scattering in the added heavy water
2) Decreased leakage due to spectrum softening
3) Increased capture due to spectrum softening ($\nu \Sigma_f/\Sigma_a$ decreases)
4) Increased capture due to reduced resonance shielding in U238.

Components 1) and 2) give positive contributions to the reactivity coefficient and components 3) and 4) give negative contributions. The present results indicate that the positive components are overestimated and/or the negative ones are underestimated. At this point it

*) An increase of the number of energy groups from 16 to 35 produced no significant change in the computed result.

may be pointed out that DTF-4 predicts a somewhat lower leakage-to-absorption ratio than MONDAY and consequently a lower reactivity coefficient, which is in slightly better agreement with experiment.

In order to achieve complete agreement with experiment the cross sections of heavy water must be reduced by about 20 % or some other cross sections of the core materials must be changed in order to shift the balance between leakage and absorption. As a somewhat arbitrary sensitivity test this balance was changed by increasing the macroscopic fission and capture cross sections by 10 % in four successive energy intervals. The result is shown in Table 5.

Table 5: Effect of cross section changes on k_{eff} and reactivity coefficient of heavy water

10 % increase in Σ_f and Σ_c in energy interval:	k_{eff}	Reactivity coefficient of D_2O divided by experimental value
No change	1.007	1.23
10^4 keV - 500 keV	1.025	1.14
500 " - 25 "	1.017	1.15
25 " - 0.4 "	1.009	1.24
0.4 " - 0 "	1.007	1.23

When the changes are applied to the higher energy intervals the leakage probability is substantially reduced and therefore components 1) and 2) of the reactivity coefficient decrease. The improvement obtained in the latter quantity is unfortunately offset by the simultaneously increased error in k_{eff}.

A test relating to the effect of anisotropic scattering was also made. The test involved a change in $\bar{\mu}$ for deuterium so as to make scattering in this material isotropic in the centre of mass system.

The corresponding change in the diffusion constant of the homogeneous core medium was of the order of 5 % at energies above 0.1 MeV. The resulting change in the reactivity coefficient of heavy water was only 1 %, however.

Finally, it may be noted that the reactivity coefficient calculated for the annular zones (Figure 5) is overestimated by approximately the same fraction (about 20 %) in all parts of the core.

The complicated dependence of the reactivity coefficient on cross sections in various energy regions renders a more precise interpretation of the observed discrepancies extremely involved. This is emphasised by the similar calculations of the effect of polythene in core 7 which, in contrast to the present results, yielded too small values of the reactivity coefficient [4].

6. SUMMARY AND CONCLUSIONS

The results obtained in the present report can be summarized in a few points:

1. The critical mass of the heavy water diluted assembly is well predicted by means of current cross section data and computer programmes.

2. The effect of the heterogeneous core composition on reactivity - or critical mass - is small. The reactivity change measured in a separate bunching experiment is (0.67 ± 0.04) %, as compared with a calculated value of 0.51 %. The latter involves several effects which are of equal order of magnitude but have opposite signs.

3. The measured reactivity coefficient of heavy water is about 20 % lower than the value obtained from calculations with the SPENG and DTF-4 programmes. Some changes in cross sections have been made in order to study the sensitivity of the calculated reactivity coefficient to various data and to improve the agreement with experiment. The improvement obtained is marginal, however, or is offset by an increased error in k_{eff}.

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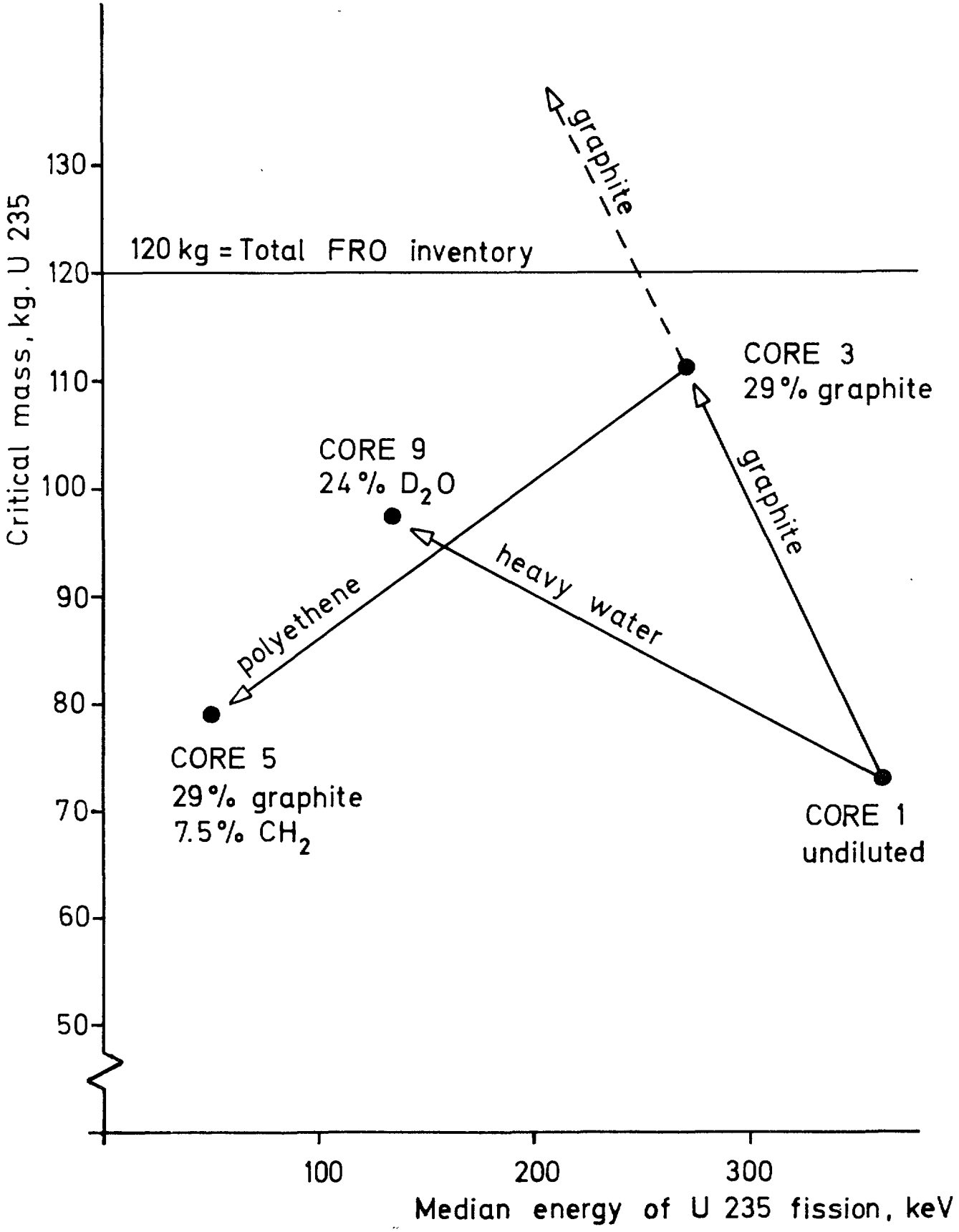

FIGURE 1: Relation between critical mass and spectrum parameter for some FRO core compositions

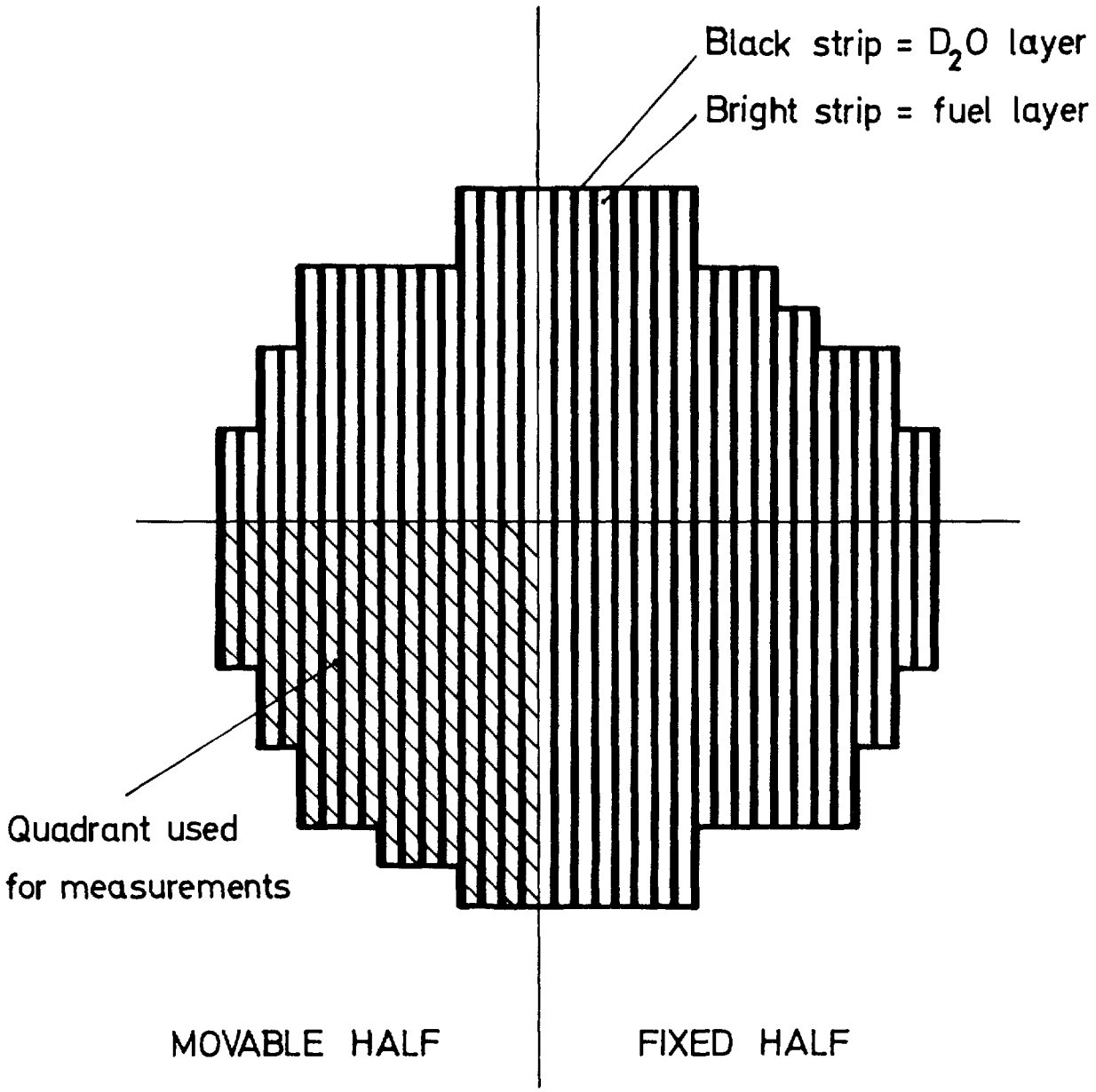

FIGURE 2 : Pattern of fuel and D_2O layers in FRO core 9

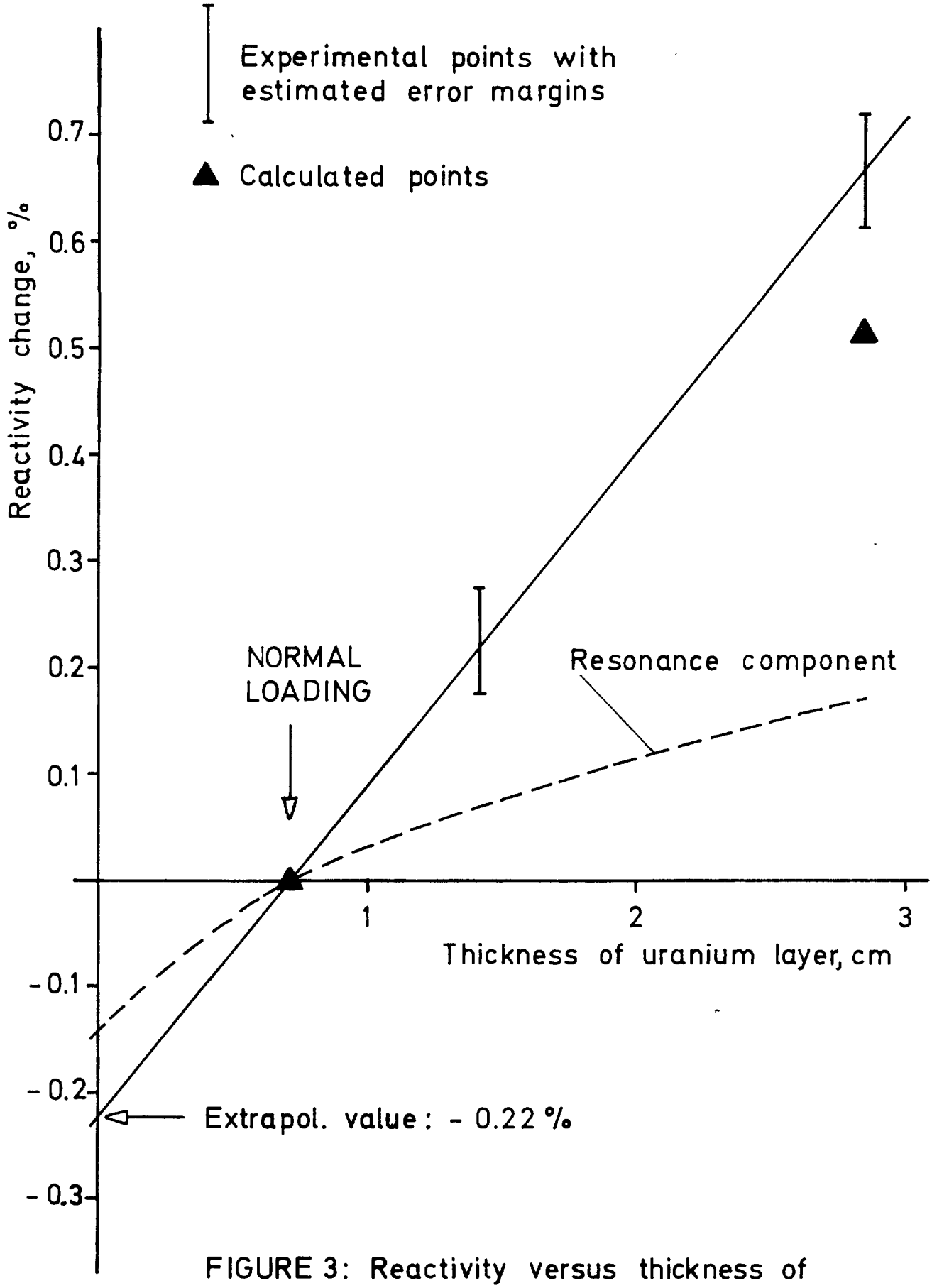

FIGURE 3: Reactivity versus thickness of fuel layer in FR0 core 9.

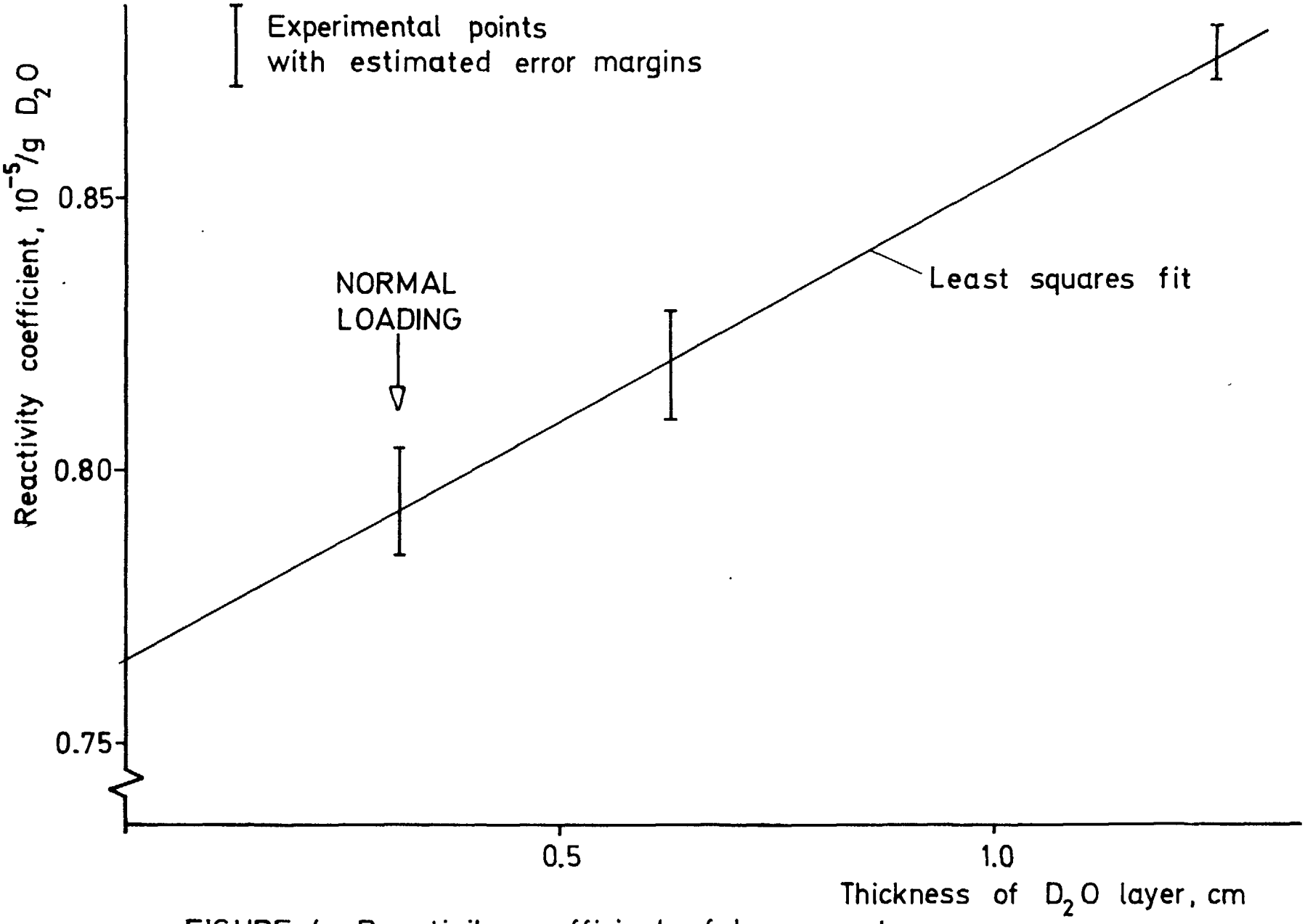

FIGURE 4: Reactivity coefficient of heavy water versus layer thickness

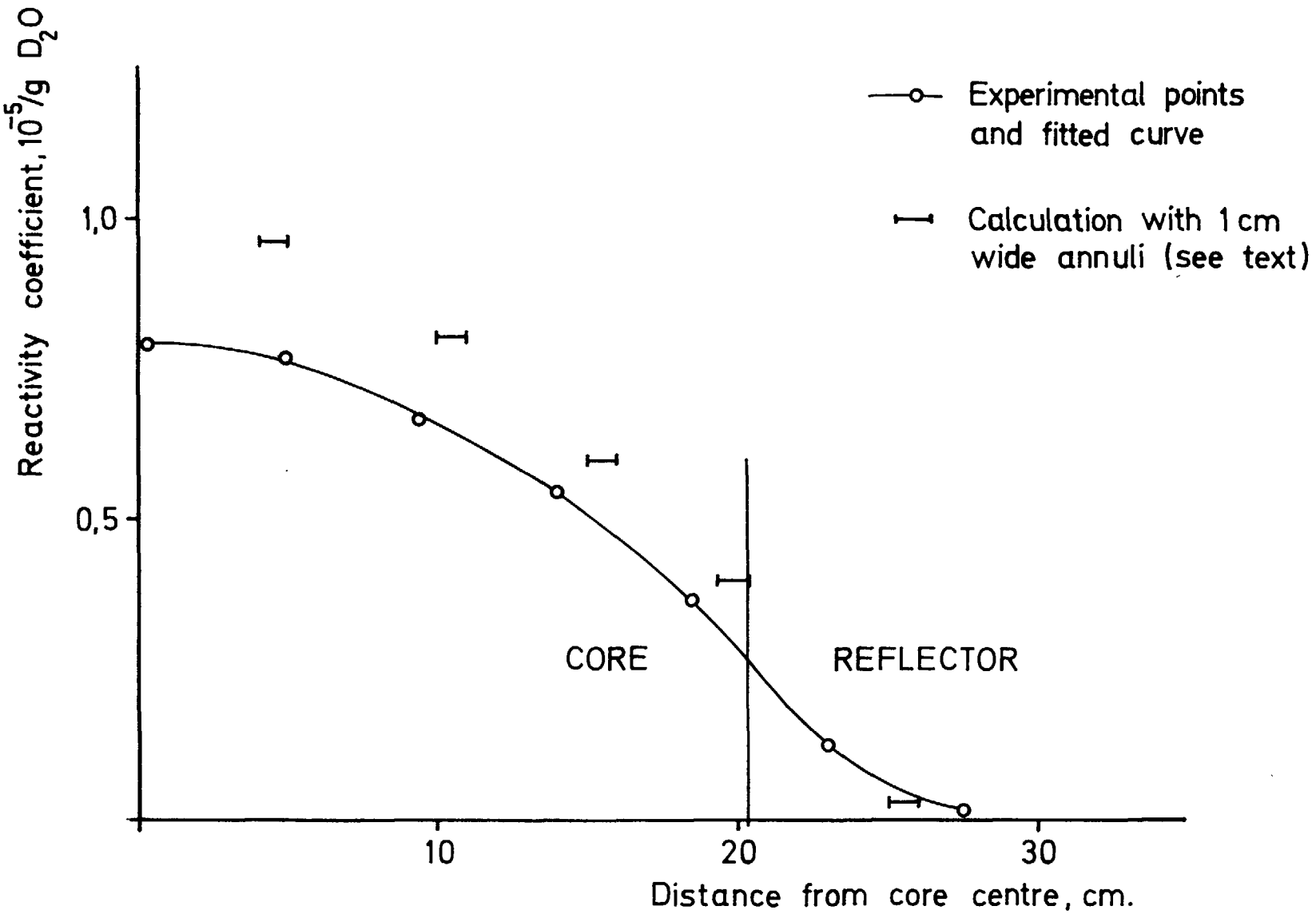

FIGURE 5: Reactivity coefficient of heavy water, radial distribution.

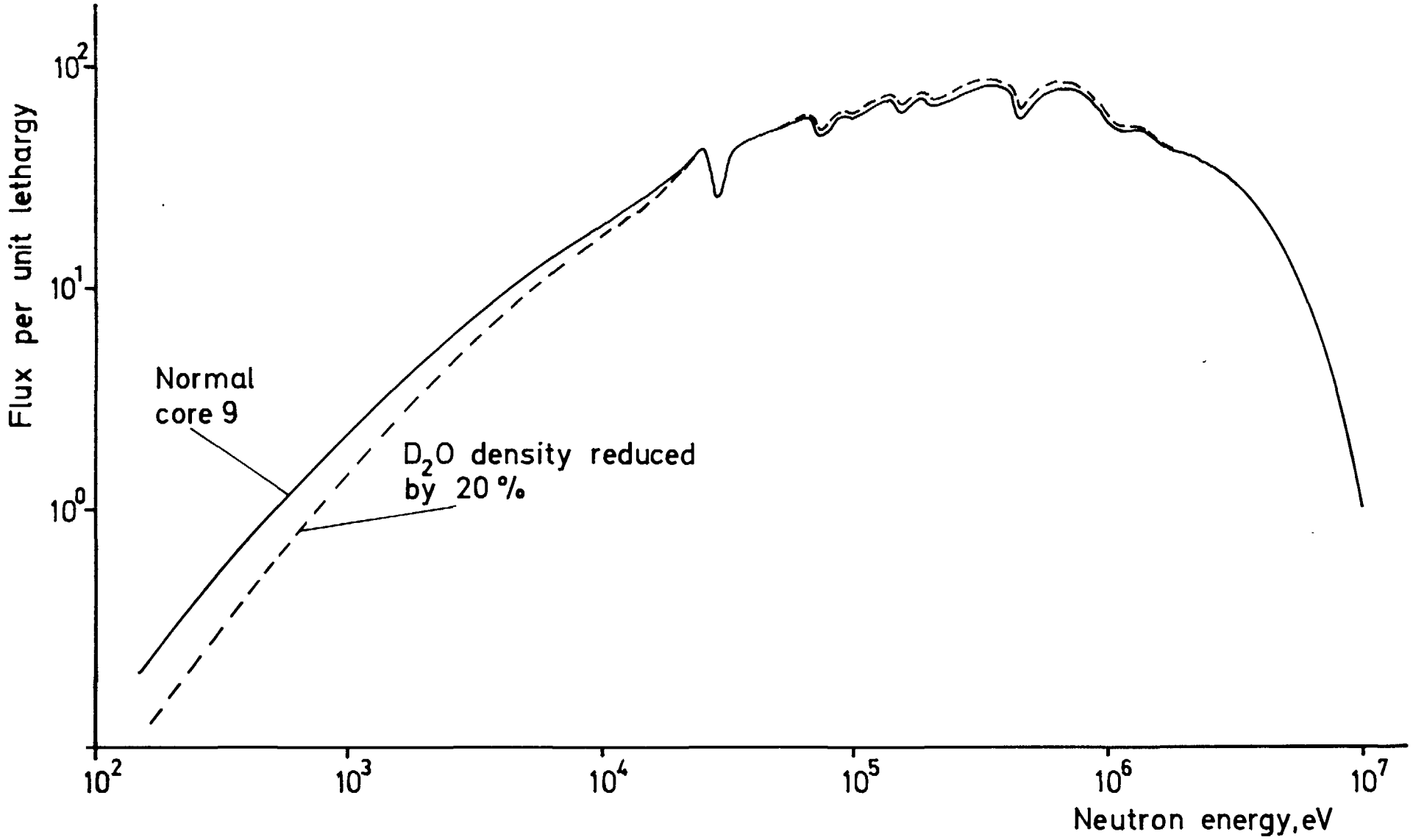

FIGURE 6: Neutron energy spectrum in FRO core 9

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