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GRAPHITE REACTOR PHYSICS

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Introduction :

1° - The study of the physics of natural uranium-graphite power reactors, initiated in France ten years ago when the Marcoule reactors were built, has been pursued along with the development of this type of reactor.

2° - The first goal of this study is the best possible knowledge of the lattice properties (k_∞ , buckling) in the different operating conditions of these reactors : cold, at full-power, during their evolution. This brought about an important experimental program using the critical assembly MARIUS [1] and taking advantage of the start-up of the first power-stations (G2, G3, EDF 1) [2] [3] [4], and advanced theoretical studies in neutron thermalization.

3° - The operation of graphite power reactors is a second subject of very general interest. The effect of the control rods both on reactivity and on flux deformations should be predicted. Numerous experiments were performed in MARIUS and EDF 1 with this in mind. On the other hand, the moderator temperature-coefficient quickly becomes positive as plutonium builds up in the fuel, which is important both in stationary operations, because of the risk of spatial instabilities, and during transients, because of the important reactivity variations concerned

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1) including work by Jean Louis CAILLY, Michel LOTT, Antoine MEYER-HEINE, Gérard TAICLET, André TESK DU BAILLER, C.E.A, et François MINNARD, EDF.

4° - Lastly, in order to predict the behavior of graphite under irradiation (Wigner effect), we needed to know the fast neutron spectrum at different points of the assembly and the neutron sources at the core and reflector boundaries.

Properties of Graphite Lattices

5° - The MARIUS critical assembly, in operation since 1960, was designed for exploration of a large domain through the variation of the lattice pitches and channel diameters ; this was made possible through the lattice changeability in a part of the reactor and through the possible use of sleeves of various diameters. The method of progressive replacement [5], successfully developed in the Commissariat à l'Energie Atomique, allowed for the study of a large number of fuel elements, dispensing with the need for excessively large quantities of Uranium. Table no. 1 shows the principal lattices studied and indicates the channel diameters corresponding to each pitch and fuel element. We systematically measured the bucklings of these lattices [6], and, in a number of cases, the fine flux structure in the cell and the spectrum indices were also measured : ^{239}Pu/^{235}U, ^{115}In/^{55}Mn, ^{151}Eu/^{55}Mn, ^{176}Lu/^{55}Mn [7].

6° - The first experiments were concerned with small pitches and small diameter fuel elements. They allowed for the development of a precise lattice computation method in the characteristic range of the first power-producing stations. The thermal utilization factor was computed with the Amouyal-Benoist method [8], the fast-fission factor with a two-group method derived from that of Spinrad (and fitted to experimental results), the migration area with the Benoist method [10], so that it was possible to fit the effective resonance integral and η on the whole set of experimental results.

7° - Actually, this method only applies when the vacuum ratio in the cell is small, for any mistake in the vacuum effect correction directly involves (k_∞ - 1). However, once a fit has been achieved with small channel lattices, we may accept as correct the values of k_∞ computed for the same lattices, but with large channels. Assuming that the migration area in the direction of the channels ($M^2_{//}$) is computed correctly, the migration area in the direction perpendicular to the channels can be deduced. Table no. 2 sums this up, and compares the values of the migration area in the perpendicular direction ($M^2_\perp$) as computed and as drawn from experiments. The generally good fit between experiment and the Benoist theory, even in cases with a very large anisotropy will be noticed.

8° - Experiments concerning large fuel elements and large pitches showed important deviations from predictions (1 to 1, 5 % on k_{∞} in extreme cases). Part of these deviations can be attributed to the computation of f and may be corrected if the neutron density ratio for uranium and moderator is calculated as a function of energy which is then averaged over the spectrum. These corrections, however, are not sufficient to account for the observed deviations. Naudet has shown [11] that the effective resonance interval should be augmented, in large cells, so as to take into account the non-uniform repartition of neutrons above 1 keV. Nor is the low energy (a few eV) neutron repartition uniform, which implies a reduction of the effective integral for large fuel elements. Generally speaking it is thus necessary, in order to perform the computation of these lattices, to look further than before into the spatial distribution of neutrons at different energies.

Plutonium containing fuels

9° - The effective fission and absorbtion cross-sections of Pu 239 are very sensitive to the shape of the spectrum in the low epithermal range, because of that isotope's resonance at 0. 3 eV. The importance of this isotope in the neutron balance of graphite reactors during fuel irradiation - half the fissions coming from plutonium at 3000 MWd/T - makes a correct knowledge of the spectrum in that energy-range necessary.

10° - Thermalization studies conducted in the CEA [12] give a representation of this spectrum by models which take into account all the important properties of the moderator and yet remain quite simple to use. Numerous spectrum indices measurements were performed in MARIUS, particularly with uranium and plutonium detectors, in order to check the calculations. The measurement of the activities of these detectors in a thermal flux on the one hand and in the studied lattice on the other, directly yield the effective fission cross-section ratio of these two isotopes as compared to that same ratio in a Maxwellian flux, and also directly yield the quantities which are needed for an evolution computation. Figure no. 1 shows the results of these measurements in different lattices characterized by $\Gamma = \frac{\Sigma_a}{\xi \Sigma_s}$) as compared to the heavy gas model computed values, and to those values computed with a more precise model : this last model takes into account both the apparent atomic mass increase of the moderator due to the crystalline bindings and the fact that the neutron looses a finite quantity of energy in a collision. This model is seen to be in excellent agreement with the experimental results.

11° - Global experiments with plutonium containingfuels are in progress in MARIUS. A progressive replacement experiment has been completed with a fuel containing 0.043 ‰ plutonium (ø 29.2) and two other experiments with higher percentages of Pu 239 and Pu 240 will be undertaken. On the other hand, an oscillation method was developed [13], which allows the simultaneous measurement of the total fission and absorbption cross sections of a sample and permits a more detailed exploration of the range we are interested in at not too high a cost.

Reactivity Evolution during Irradiation [14] :

12° - Reactivity evolution during irradiation results from a compromise between the destruction of ^{235}U and the production of ^{239}Pu. Since the two phenomena are of the same order of magnitude, the result is specially sensitive both to the initial conversion rate and to the effective cross-sections of plutonium 239. The evolution curves are thus very dependant on the moderator temperature as is shown in figure no. 2 where the punctual evolution curves of EDF 1, EDF 2 and EDF 3 are plotted. The effective cross-section of ^{240}Pu, even though distinctly not as well known, is less important, because the larger or smaller amount of Pu 241 produced balances the larger or smaller absorbtion by ^{240}Pu. Figure no. 2 shows that, exclusively from the neutronics point of view, a very high burnup can be achieved. In the case of continuous and uniform fuel circulation, the maximum irradiation is given by the condition of equal areas above and below the abcissa axis, that is, about 6000 MWd/T for EDF 3. In practice, a realistic fuel-cycle scheme should allow the achievement of a 4500 MWd/T minimum burn-up, a 1 ‰ reactivity reserve being conserved.

13° - The formation of plutonium makes for a fast increase in the moderator temperature coefficient (figure no. 3). This coefficient is, of course, dependant on the quantity of plutonium produced and on the temperature. The computations are very dependant on the slowing-down model adopted : for a 2000 MWd/T irradiation at 350°C, α_M changes from a value of 8 pcm/°C using the Westcott effective cross-sections (1958) to 13 pcm/°C using the models presently exploited at Saclay.

14° - The complexity of the phenomena related to fuel evolution makes it necessary that the calculation methods be checked against the very results of irradiation experiments.

15° - An important program was thus accomplished in G3 [3], which will be developed in EDF 1 in the first years of its operation. On the one hand, the evolution of reactivity is measured by means of the control rod motions which become necessary. On the other hand, the measurement of the moderator and uranium temperature coefficients was made possible by the study of the dynamic behaviour of the reactor following a perturbation (variation of the inlet temperature, insertion of antireactivity, etc...). The results of the experiments made so far are in close agreement with predictions.

16° - A more detailed analysis of the irradiation effects is, however, desirable ; it should distinguish between the evolution of the chemical and isotopic composition of the fuels and the effects on neutron balance of the modifications of that composition. This is achieved through the combination of oscillating irradiated fuel elements and chemical and physical analysis of these fuel elements ' composition. We thus have at our disposal fuel elements from G2 and G3 for oscillation experiments in MARIUS with burn-ups over 2500 MWd/T. Some 40 fuel elements also have been inserted in EDF 1 and more will be in EDF 2 after a careful intercalibration.

17° - The above paragraphs have emphasized the influence of temperature on all the phenomena related to the existence of plutonium in graphite reactors. This is why the Commissariat à l'Energie Atomique and Electricité de France decided to construct in Cadarache a critical assembly, CESAR, which can operate at temperatures up to 500°C. CESAR, which will be completed this year, will be used in the same type of experiments as MARIUS : buckling measurements by the flux-map and progressive replacement methods, fuel oscillations. The latter, as in MARIUS, will be used especially with irradiated fuel elements.

Control Rods

18° - The use of control rods of various shapes and efficiencies in order to answer different demands - piloting, compensating, safety - in addition to the desire for the acquisition of a good knowledge of their efficiency, led to the development of a precise method of computation. The extrapolation length is computed, on the boundary of the channel which contains the control rod, as a function of energy, and the inverse of this extrapolation length is averaged first on the thermal part of the spectrum, then on the slowing-down neutron spectrum ;

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an upper limit is chosen for this last integration. Extrapolation lengths for the two-group diffusion computations are thus obtained, their definition bling cohérent with that of the lattice diffusion coefficients.

19° - Experiments were made in MARIUS, which were either measurements in critical configurations or subcritical pulsed-neutrons measurements. The control rod diameter was systematically changed (from 35 to 100 mm), as were the diameter of the rod-containing channel, the vacuum ratio and the moderating ratio of the lattice surrounding the rod. There was excellent agreement between experiments and computations (better than 3 ‰), provided that the transport mean-free-path used to compute the extrapolation lengths was that of the lattice and not of the moderator, and that the upper limit of the fast-group integral was fitted to an energy well above 2 MeV.

20° - The application of these results to the practical cases of power reactors is achieved through the use of two or three-dimensional computation programs and presents no basic problem. Numerical difficulties encountered when the mesh step changes too rapidly led us, however, to determine from the above computations values of the extrapolation lengths which can be used on a different boundary or to determine constants for a large enough region which would absorb as many neutrons, in each group, as the control-rod. Both methods led to results which are quite comparable to those drawn from experiments in EDF 1.

Dynamic behavior of power reactors

21° - In a power reactor, the multiplication coefficient, k, is, at a given point, a function of the flux at that same point, because of the reactivity coefficients associated to the uranium temperature (α_U), the moderator temperature (α_M) and the xenon concentration (α_X). In the natural uranium, graphite-gas reactors, these reactivity coefficients are of the following orders of magnitude : α_U = - 2 pcm/°C, α_M from - 3 pcm/°C to + 15 pcm/°C as a function of fuel burn-up, α_X = 3100 pcm.

22° - A local flux variation entails :

- a variation of temperatures in the same direction, hence a variation of k in the same direction or in an opposite direction depending on the sign of the temperature coefficient.

- a variation in the opposite direction of the xenon concentration (since there is an immediate variation of the destruction rate, whereas the creation rate has a time constant of many hours), hence a variation of k in the same direction, the importance of which increases with specific power.

- a variation in the same direction in neutron leakage, the importance of which decreases when the migration-area to reactor dimension ratio decreases.

23° - There are thus two stabilizing effects : uranium temperature and neutron diffusion ; one destabilizing effect : xenon concentration ; one stabilizing or destabilizing effect : the moderator temperature, depending on the sign of α_M, which, however, intervenes after a delay of some minutes because of the heat capacity of graphite. The superposition of these various effects leads in certain cases to a spatial flux instability, despite the fact that the piloting system keeps the total power production constant.

24° - A simple analytical formulation [15] which neglects the time constants associated with the variations of the flux distribution and of the uranium temperature as compared with those associated with the moderator temperature and with the kinetics iodine and xenon, shows the existence of a value of α_M associated to each flux harmonic, beyond which that harmonic, if excited, will amplify with time. The application of this method to EDF 3 shows that excitation of the first azimuthal harmonic (i. e., increase in the flux in one half of the reactor and decrease of the flux in the other) provokes an oscillating response which converges for $\alpha_M < 3$ pcm/°C and diverges for $3 < \alpha_M < 7$ pcm/°C, and a purely exponential response, with a doubling time which may be as small as 15 minutes for $\alpha_M > 7$ pcm/°C. The corresponding values of α_M for the higher order harmonics are somewhat larger (because the stabilizing leakage effect is greater), as they are for the axial harmonics because the transport of energy due to the coolant is a stabilizing factor on the temperature-coefficients.

25° - The study of the control of these instabilities is much more complex, and demanded the development of two-dimensional kinetics computation program (r- θ and r-z geometries) [15] . These programs allowed us to check the results of the simplified analytical study, and to define more precisely both the location and efficiency of the spatial piloting rods needed to overcome that phenomenon and the practical use of these rods.

26° - A second consequence of the high moderator temperature coefficient is the important reactivity variation between the reactor when operating at full power and when stopped. For instance, during the start-up, reactivity will at first increase 2 ‰ to 3 ‰, and will then decrease as xenon builds up. One must provide a large number of control rods, the individual efficiency of each one being small enough so as not to provoke too large flux perturbations.

Fast and Intermediary Fluxes :

27° - Knowledge of the fast neutron fluxes in a graphite reactor is related to the problem of the evolution of the physical properties of the moderator under irradiation and especially to the deformations which give birth to stresses within the assembly ; this knowledge is also related to the calculation of neutron leakage in reflectors with holes. These problems are particularly important in the advanced graphite natural uranium projects [18] where the lattice pitches are large and the channel diameter can reach 180 mm.

28° - Numerous measurements were made in MARIUS for 224 and 317 lattice pitches and in EDF 1 ; we had in view both a better fundamental understanding of the fast flux [16], and a comparison of the graphite irradiations in EDF 1 with those in the other reactors of the Commissariat à l'Energie Atomique [17].

29° - Figure no. 4 shows the flux distribution measured with rhodium detectors (40 keV threshold) and phosphorus detectors (1.5 MeV threshold) for a 50 mm diameter full rod, a 110 mm channel, with a square lattice of 317 mm pitch. The results are expressed in equivalent fission flux.

30° - These experimental results may be reconstructed if the flux is analysed in an homogeneous and an heterogeneous component. The homogeneous component may be computed starting from fictitious sources assumed to have a uniform repartition in the moderator. Its value is approximately 10^{12} P, P being the specific power in watts per cubic centimeter of moderator. The heterogeneous component is connected to neutrons which have undergone few collisions and can be treated in a first approximation as a no-collision flux of neutrons born in the fuel rods.

Conclusion.

31° - From this complex set of theoretical and experimental studies, three main conclusions may be drawn :

- the very broad exploration of the graphite lattices which became possible from 1960 on due to the MARIUS critical assembly paved the way for an important evolution in reactor projects, allowing as it did the use of large fuel elements. MARIUS will be transformed towards the end of 1964 in order to facilitate the study of those types of lattices.

- The plutonium-containing lattice research program, which has demanded considerable investments in various domains, has already assured us that, at least from the neutronic viewpoint, high burn-up rates can be achieved in natural uranium graphite reactors.

- the fundamental knowledge obtained these last years, and that which will be drawn from the continuing research, will enable the physicists to answer the needs of the plant operators so as to draw maximum profit from the large power stations now under construction.

TABLE N° 1

Lattices studied in the MARIUS critical assembly

Fuel element (mm)	Uranium cross section (cm^2)	Lattice pitch (mm)				
		192	224	271,5	316,8	384
28	6	70, 90, 110	70			
29,2 [1]	6,7		70			
31	7,4	70	70, 90, 110, 140			
40	12,4		70	70	70, 90	
50	20		70	70	70, 90, 110	140
64	29				90	
10,7 x 32,7 (T-1)	7,5		70			
20,6 x 40 (T-2)	9,3	70, 90, 110	70			
29,5 x 45 (T-3)	9,5	70	70			
35 x 45 (T-4)	6,3	90	70			
30 x 50 (T-5)	12,5		70	70	70, 90	
46 x 64 (T-6)	15,5				90	
65 x 82 (T-7)	20				90, 110	140
90 x 103 (T-8)	20				110	140
$T_6 + T_4$	22				90	
$T_7 + T_4$	26				90, 110	140
$T_8 + T_4$	26				110	140
$T_7 + T_6$	35				90	140
$T_8 + T_6$	35					140
EDF 1	8	90	90			
EDF 2	10		70	70		

1) Various sets of fuel-elements are at hend : natural uranium, 0,69 % depleted uranium, 0,83 an 0,86 % enriched uranium, uranium and plutonium.

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TABLE II.

Lattice	Channel	$B^2_\perp$ (m^{-2})	$B^2_{/\!/}$ (m^{-2})	K calculated	$M^2_{/\!/}$ calculated (cm^2)	$M^2_\perp$ calculated (cm^2)	$M^2_\perp$ measured (cm^2) 2)
Pitch 192 Ø 28	70	0,903	0,277	1,08724	759	708	711 ± 17
	90	0,722	0,2575	1,08566	962	823	821 ± 30
	110	0,551	0,240	1,08050	1298	999	861 ± 55
Pitch 224 Ø 31	70	0,940	0,293	1,09030	754	719	714 ± 8
	90	0,843	0,281	1,09314	894	797	787 ± 20
	110	0,568	0,358	1,09353	1134	925	898 ± 54
	140	0,363	0,222	1,08775	1771	1242	1305 ± 53
Pitch 317 Ø 40	70	0,357	0,299	1,05583	839	826	840 ± 17
	90	0,444	0,291	1,06589	892	851	878 ± 30

2) The errors quoted relate to the experimental errors on B^2 and B^2

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TABLE III.

Lattice	Element	(φ_u/φ_m) A. B.	$(\varphi_u/\varphi_m) = \bar{h}$	(φ_u/φ_m) experimental	$\frac{\text{exp.} - \text{A. B}}{\text{A. B.}}$ %	$\frac{\text{exp.} - \bar{h}}{h}$ (%)
Pitch 224	Ø 31	0, 522	0, 531	0, 539	3, 3	1, 6
Channel 70	Ø 50	0, 362	0, 413	0, 410	11, 6	- 0, 7
Pitch 316, 8	Ø 40	0, 409	0, 416	0, 417	2	0, 2
Channel 70	Ø 50	0, 324	0, 344	0, 351	8, 3	1, 8

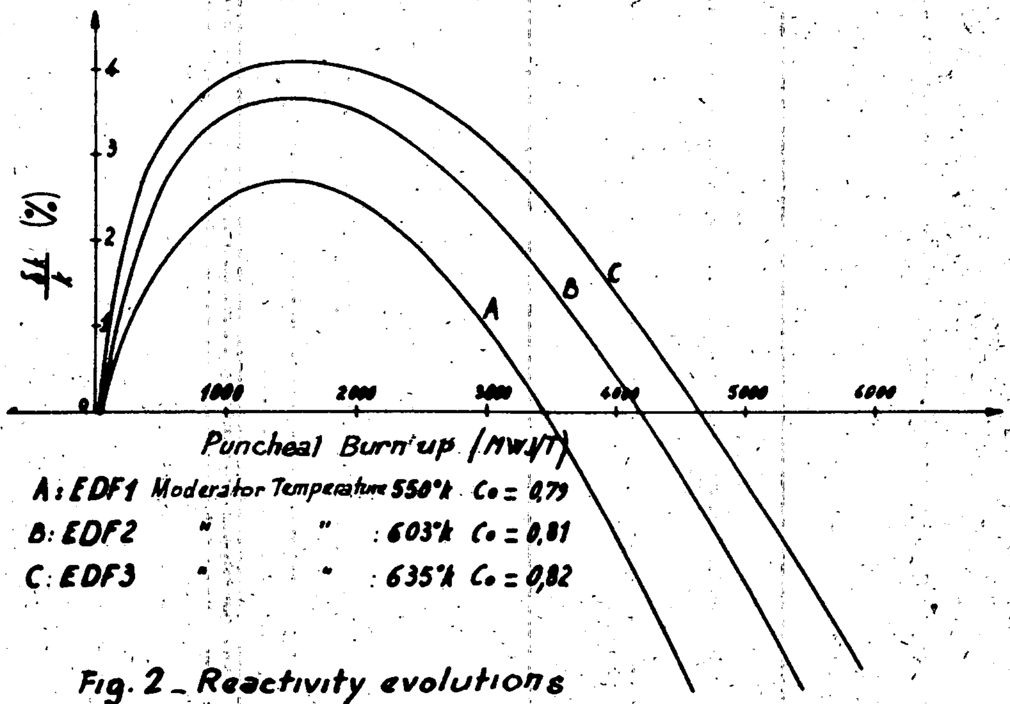

Fig. 2 _ Reactivity evolutions

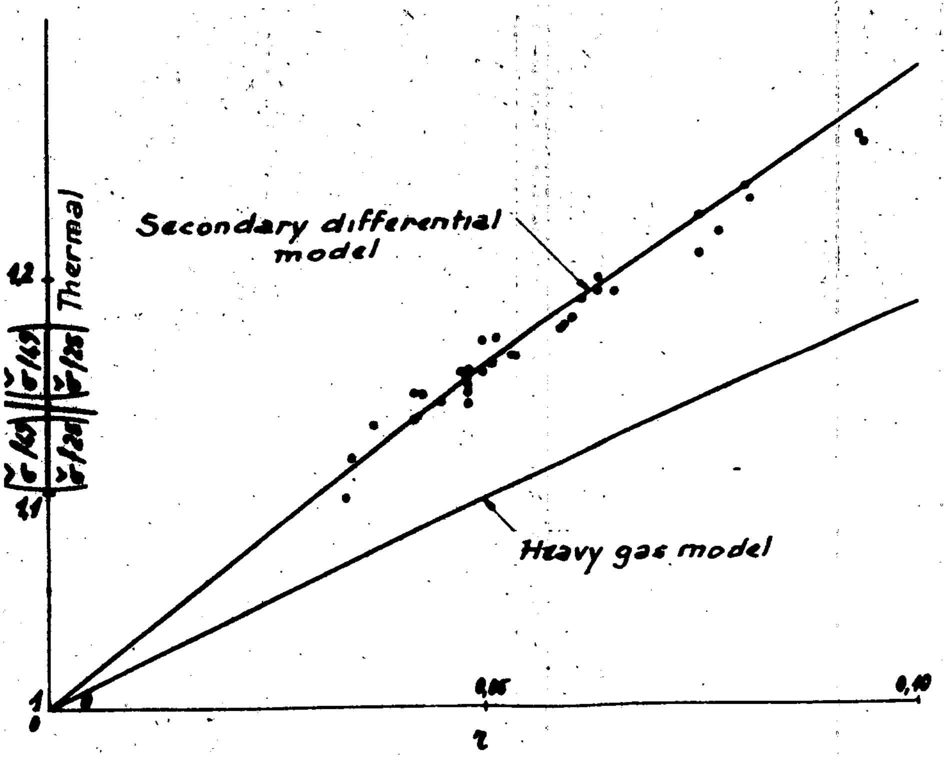

Fig. 1 - Spectral (PU/U) in moderator

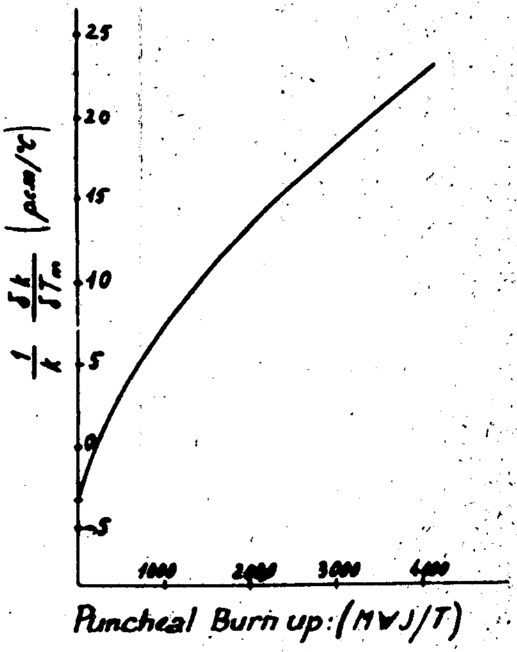

Fig. 3 - Moderator Temperature coefficient

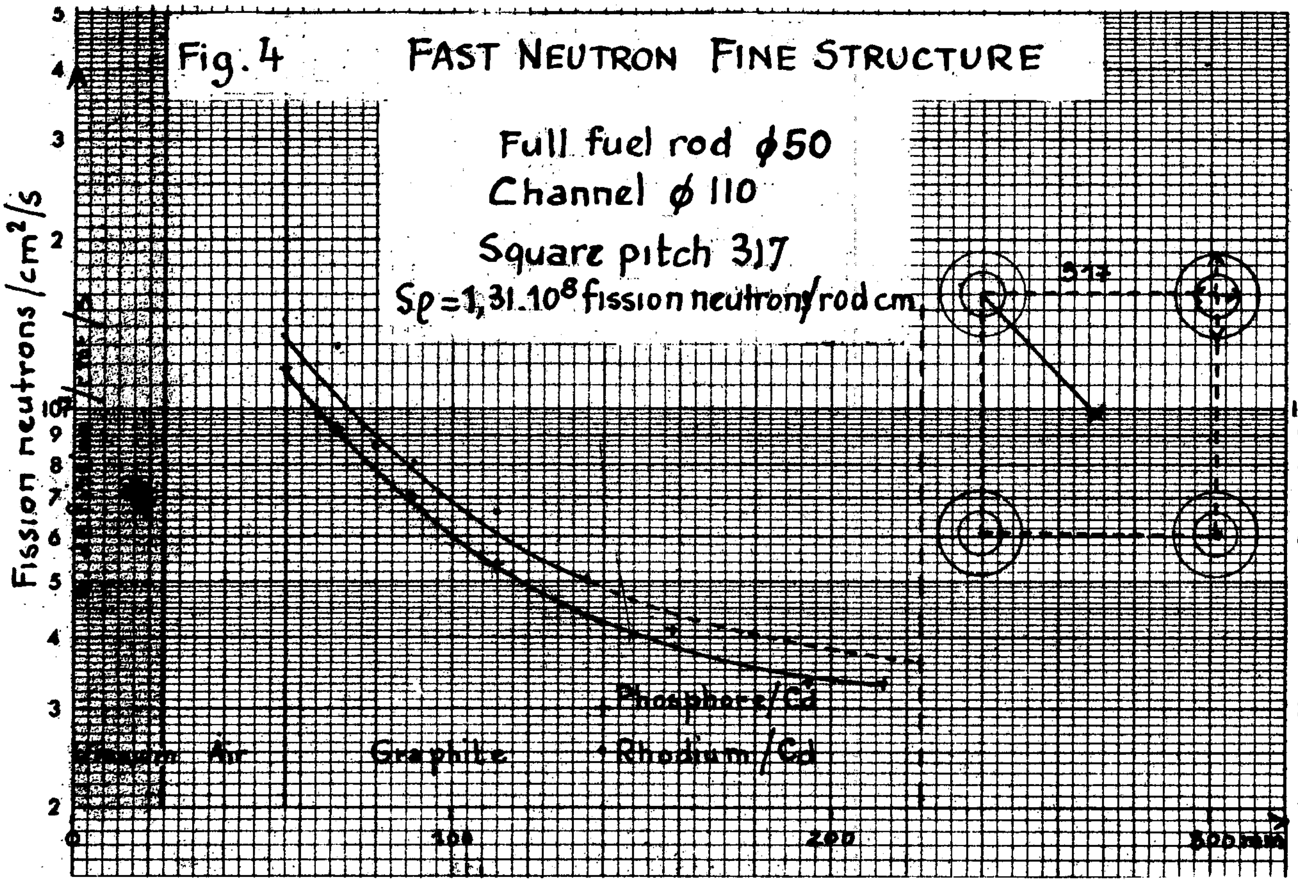

Fig. 4 FAST NEUTRON FINE STRUCTURE