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CRITICAL EXPERIMENTS ON NATURAL URANIUM OXIDE, ORGANIC COOLED, HEAVY WATER MODERATED LATTICES

by

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A series of critical experiments has been carried out at Saclay, France, by the Reactor Physics Department of the C.C.R., Euratom. The purpose of the measurements was to determine the material buckling of nine different ORGEL lattices using natural uranium oxide fuel elements, moderated by heavy water and cooled by an organic liquid.
This report describes the experimental technique, the results obtained and their comparison with calculated values. The latter turn out to be systematically lower than the former. Some criteria are given to improve the agreement between the two sets of values.

Critical experiments on natural uranium oxide, organic cooled, heavy water moderated lattices

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1. INTRODUCTION

A set of experiments was carried out during the months of June and July, 1961, on the Aquilon II heavy water-natural uranium research reactor, at Saclay, France. These experiments were programmed and performed by the Reactor Physics Department of the C.C.R. Ispra, Italy, in cooperation with the personnel of the French « Commissariat à l'Energie Atomique ».
The object of the measurements was to determine the material buckling of natural uranium oxide lattices, moderated by heavy water and cooled by organic liquid, of the type now being studied and worked on by Euratom under the ORGEL (Organique Eau Lourde) research program.
The measurements were taken using a method worked out by R. Naudet and co-workers at Saclay [1, 2] which consists in the progressive substitution of the elements located in the central zone of the reactor with the elements to be tested. This method allows the accurate determination of a lattice material buckling when a limited number of elements, insufficient to make up a critical reactor, is available.
In the experiments performed by the authors the reactor was filled with 112 seven-rod clusters

(reference elements) directly placed in the heavy water. The elements of the ORGEL type to be tested were 19-rod clusters of natural uranium oxide plunged into organic liquid and contained in an hexagonal aluminium tube.
Three buckling measurements by flux mapping were made on the reference lattice, at 19, 21 and 24 cm pitches respectively. Afterwards the substitution measurements were made for each of the three pitches.

2. DESCRIPTION OF FUEL ELEMENTS

The reference fuel element characteristics were as follows (fig. 1):

— diameter of UO_2 pellets	22	mm
— can material	Al	
— can inside diameter	23	mm
— can thickness	1	mm
— UO_2 density	10.23	g/cm³
— center-to-center distance between rods	32	mm

Three types of ORGEL fuel elements having the following characteristics were used for the measurements (fig. 2):

	A0-12-0	A0-12-1	A0-16-1
UO_2 pellets diameter (mm)	12	12	16.2
UO_2 density (g/cm³)	10.15	10.15	9.85
can inside diameter (mm)	13	13	17
can thickness (mm)	1	1	1
can material	Mg	Mg	Al
organic liquid volume per unit height (cm³/cm)	12.38	12.38	17.43
equivalent inside diameter of tube (mm)	76.5	76.5	95.28
volume of hexagonal tube per unit height (cm³/cm)	2.83	2.83	3.15
center-to-center distance between rods (mm)	15	16	20

The organic liquid used was monoisopropyldiphenyl ($C_{15}H_{16}$) with a density at 20 °C of 0.973 g/cm³ (4.78×10^{22} hydrogen atoms per cm³).

A chemical analysis carried out on a sample piece of aluminium taken from a tube showed the presence of the following impurities having an appreciable effect on the aluminium absorption cross section (% by weight):

Mn = 0.01
Si = 0.18
Fe = 0.32
Cu = 0.01

3. REFERENCE LATTICE BUCKLING MEASUREMENTS

The Aquilon II reactor was filled with 112 reference elements. The equivalent radius of the core at the 19, 21 and 24 cm pitches turned out to be 113.5, 125.4, and 143.4 cm, respectively.
The container not being completely filled up radially by the 112 elements, the remaining heavy water radial reflector in the three cases was 31.5, 19.6 and 1.6 cm respectively.
Under slightly supercritical conditions (flux-doubling time of at least 100 seconds) a certain number of manganese detectors positioned in the central zone of the core were irradiated. The irradiation was carried out until an integrated flux of 10^9 n/cm² was attained in the center of the core.
The manganese detectors had the following characteristics:
foil thickness: 0.15 mm
foil diameter: 6.15 mm

The detectors were fitted in 2 m long aluminium tubes having an inside diameter of 7.5 mm and an outside diameter of 9 mm. Their position along the tube was determined within 1 mm.
The tubes were hung up vertically inside the core with the lowest detectors located 10 cm above the bottom of the core. They were placed each in the center of the square cell formed by 4 contiguous elements. The cells were chosen so as to obtain the lining up of the tubes.
From 4 to 10 detectors were placed in each tube. During the irradiation all the tubes contained the same number of detectors, placed on different horizontal planes. The determination of each buckling was carried out by repeating the irradiation seven times, for a total of 280 detectors irradia-

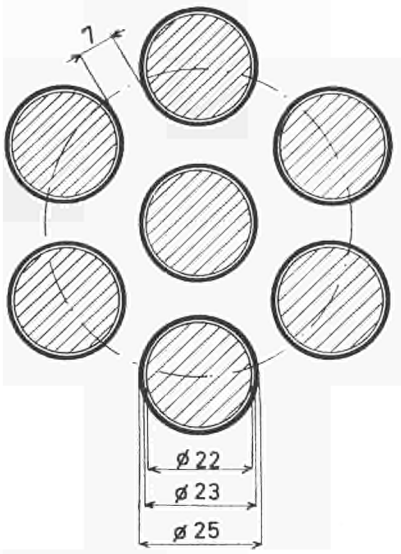

Fig. 1 - Reference fuel element.

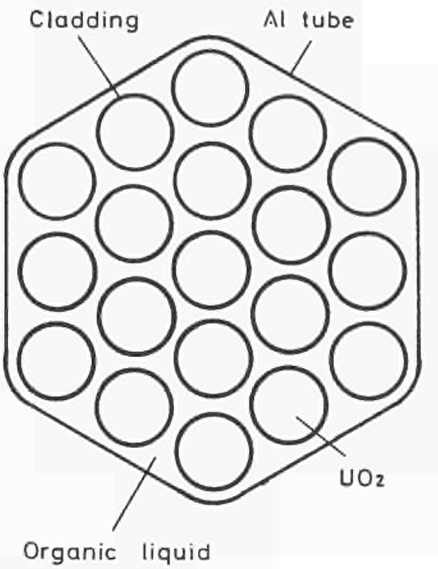

Fig. 2 - ORGEL type fuel element.

ted. Approximately 250 of these were found to be of use for flux mapping.

The activity of the detectors was then measured and the results of the measurements were fitted to the theoretical shape of the flux in the core. The flux at the center of the core, where the distorting effect introduced by the reflectors can be assumed to be negligible, can be described by the function:

$$\phi(r, z) = A\, J_0(\beta r) \sin[\alpha(z - \gamma)]$$

The corrections due to the detectors and the counting equipment were carried out using a special code for digital computer. From the results of counting one value of the coefficients A, α, γ, was found for each tube and a value of β for each level of detectors. Program SPM/001 on IBM 7090 was used for this purpose.

The series of values of α and β thus obtained was first averaged out arithmetically and then by using the inverse square of the deviation as weight.

Afterwards, the value of α was corrected; in fact the moderator level during irradiation was higher than in critical conditions when the detectors and ionization chambers are extracted from the reactor. Therefore, the difference between the irradiation level and the level determined by kinetic approach, as well as the height of water equivalent to the anti-reactivity of the chambers were subtracted from the extrapolated height H_e.

The uncertainty on the values of α and β was calculated for a probability of 95% and 99%.

The processing and working out of the results of the measurements following the method described was done at Saclay by Mr. Chabrillac [3, 4]. The following figures were obtained:

Pitch (cm)	19		21		24	
H_e (mm)	1775.9	$\pm$ 7 $\pm$ 10	1669.4	$\pm$ 8 $\pm$ 11	1714.0	$\pm$ 13 $\pm$ 17
α (m^{-1})	1.769	$\pm$ 0.007 $\pm$ 0.010	1.882	$\pm$ 0.009 $\pm$ 0.012	1.833	$\pm$ 0.018 $\pm$ 0.014
R_e (mm)	1538.9	$\pm$ 15 $\pm$ 20	1630.65	$\pm$ 22 $\pm$ 28	1705.7	$\pm$ 9 $\pm$ 12
β (m^{-1})	1.563	$\pm$ 0.016 $\pm$ 0.020	1.475	$\pm$ 0.020 $\pm$ 0.026	1.410	$\pm$ 0.008 $\pm$ 0.010
B^2_m (m^{-2})	5.572	$\pm$ 0.075 $\pm$ 0.100	5.716	$\pm$ 0.090 $\pm$ 0.120	5.348	$\pm$ 0.075 $\pm$ 0.095

The measurements taken at the various pitches were made at different heavy water isotopic concentrations; the values of B^2_m shown above were thus all brought up, as indicated in the following table, to the standard purity of 99.800%:

Pitch (cm)	Heavy water purity (%)	Correction (m^{-2})	B^2_m (m^{-2})
19	99.695	+ 0.105	5.68
21	99.700	+ 0.115	5.83
24	99.690	+ 0.150	5.50

4. SUBSTITUTION MEASUREMENTS

The experiments were carried out in the following manner.

First of all the critical level of the reactor charged with the 112 reference elements was determined by kinetic approach. The four central fuel elements were then extracted from the reference core and replaced by four ORGEL fuel elements. The critical level was measured again with the new elements inserted.

The same was done in the same way after replacing 12, 16 and 24 fuel elements (fig. 3).

In this manner five values of critical level for each type of fuel element and for each pitch (one of which values relating to the reference) were obtained. A general outlook of all these values is shown in fig. 4. For more details on experimental techniques see ref. 6.

The principle of interpretation is the following: knowing the critical dimensions of the core before and after substitution, the K_∞ and the diffusing properties of the ORGEL lattice, its material buckling can be found by means of a two group analysis.

On principle, following this method, a single substitution would be sufficient to determine the buckling of the test lattice. It must, however, be pointed out that the method of two-group analysis applied to the two multiplying regions (considered as being homogeneous), turns out to be insufficient for an evaluation of the coupling between the two zones. This interaction which, in the theory of the method of substitution, is principally described by means of a single parameter, can be determined experimentally by carrying out several substitutions.

In our experiments the substitution of four elements appeared to be useless, the corresponding results not being in agreement with the results of the other three replacement measurements.

This anomaly appeared for practically all the lattices taken into consideration and was evidently due to the fact that the hypothesis of homogenization is not applicable to a region composed of four elements only.

The values found for ΔB^2 were:

Element	A0-12-0 (Mg)			A0-12-1 (Mg)			A0-16-1 (Al)		
Pitch (cm)	19	21	24	19	21	24	19	21	24
ΔB^2 (m^{-2})	— 1.71	— 2.24	— 2.31	— 1.56	— 2.00	— 2.07	— 2.77	— 2.44	— 1.79
B^2_{ref} (m^{-2})	5.68	5.83	5.50	5.68	5.83	5.50	5.68	5.83	5.50
B^2 (m^{-2})	3.97	3.59	3.19	4.12	3.83	3.43	2.91	3.39	3.71

The main sources of error in the ΔB^2 evaluation were the following:

a) uncertainty due to the experimental value of the extrapolated height H_e of the reference core. The confidence interval on H_e was calculated assuming a probability of 99%;

b) uncertainty due to the experimental value of the radial buckling of the reference core. Error calculation hypothesis assumed as for *a*) above;

c) uncertainty as to the theoretical estimate of the difference between the axial reflector savings of the two zones; it was assumed that the estimate of this difference should be affected by a maximum error of 10%;

d) uncertainty as to the ratio between the diffusion coefficients of the two zones. A maximum error of 20% in the parameter $t - 1$ (ref. 2) which characterizes the deviation of this ratio from unity, was assumed;

e) uncertainty as to the reflector coefficients e, e', e'' (ref. 2). It was assumed that there was an uncertainty of 15% in respect to these coefficients;

f) uncertainty due to insufficiency of the theory (unreliability of the criteria of homogenization when applied to small regions, failure of the two group model in border regions). It was estimated (ref. 1) that a maximum error of 2% in ΔB^2 properly takes into account such uncertainties.

On the basis of the above-given criteria the total error was evaluated for each ΔB^2 measured. It is shown in the following table:

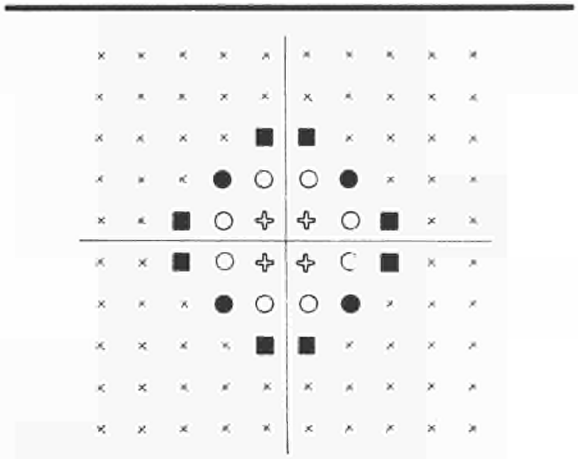

Fig. 3 - Layout of the successive substitutions.

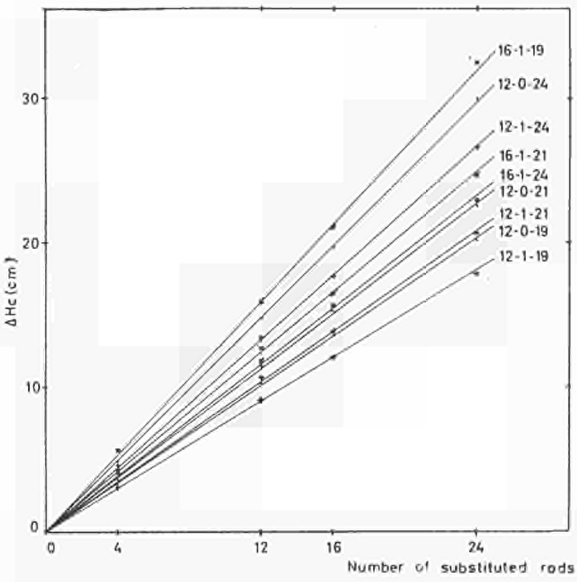

Fig. 4 - Critical levels.

Element	A0-12-0 (Mg)			A0-12-1 (Mg)			A0-16-1 (Al)		
Pitch (cm)	19	21	24	19	21	24	19	21	24
Error on ΔB^2 (m^{-2}) $\pm$	0.076	0.088	0.090	0.073	0.080	0.080	0.102	0.096	0.070
Error on B^2 (m^{-2}) $\pm$	0.125	0.149	0.130	0.124	0.144	0.124	0.143	0.153	0.118

5. NOTES ON ORGEL LATTICE CALCULATION METHOD IN USE BEFORE CARRYING OUT THE EXPERIMENTS

The bucklings obtained from measurements were compared with the values calculated by the method presently in use at the Euratom Reactor Physics Department to evaluate the reactivity balance in non-irradiated heavy water moderated, organic liquid cooled natural uranium lattices.

This method of calculation utilizes the results of the French correlation for heavy water cooled and moderated lattices, which was established on the basis of a great number of buckling measurements (nearly a hundred) made at Saclay from 1956 to date on the Aquilon reactor. The interpretation of these measurements was made on the basis of the two-group critical equation:

$$\eta\, p\, \varepsilon\, f = (1 + B^2 L^2)\ (1 + B^2 L_s^2)$$

which can be written:

$$\ln(\eta\, p) = \ln \frac{(1 + B^2 L^2)\ (1 + B^2 L_s^2)}{\varepsilon\, f}$$

L^2, L_s^2, ε, f, are computed. B^2 is the measured value. A plot is made of $y = \ln(\eta\, p)$ versus:

$$x = \frac{V_u}{(\xi\, \Sigma_s)_o\ V_o + (\xi\, \Sigma_s)_m\ V_m} P_B$$

The points obtained by varying the lattice pitch for a given fuel element lie, within a small margin of error, along a straight line. We can then write:

$$\eta\, p = A \exp(-x\, I_{\text{eff}})$$

ln A and I_{eff} are the initial ordinate and the slope of the straight line plot. Further, a single value of A can be determined for all the considered fuel elements. Once determined this value of A, for each fuel element I_{eff} can be graphically determined. The values of I_{eff} are plotted vs $\sqrt{\frac{S}{V_u}}$. The points lie, again within a small margin of error, along a straight line. We write:

$$I_{\text{eff}} = a + b \sqrt{\frac{S}{V_u}}$$

a and b can be determined as the intercept and the slope of the straight line. In the case of heavy water moderated and cooled lattices, A was found to be 1.294; for a and b two sets of values, one for metal uranium and the other for uranium oxide, were obtained.

In the case of the ORGEL type lattices, we assumed the same figures of A, a, b, as for heavy water lattices. As to the quantities to be calculated, we modified the French formulae to take into account the presence of hydrogen atoms.

6. COMPARISON BETWEEN CALCULATED AND MEASURED BUCKLING VALUES

The comparison between the measured bucklings and the values calculated using the above described method shows that the latter are systematically lower than the former, and the difference, in some cases, attains 0.5 m^{-2} (see fig. 5). The slope of the

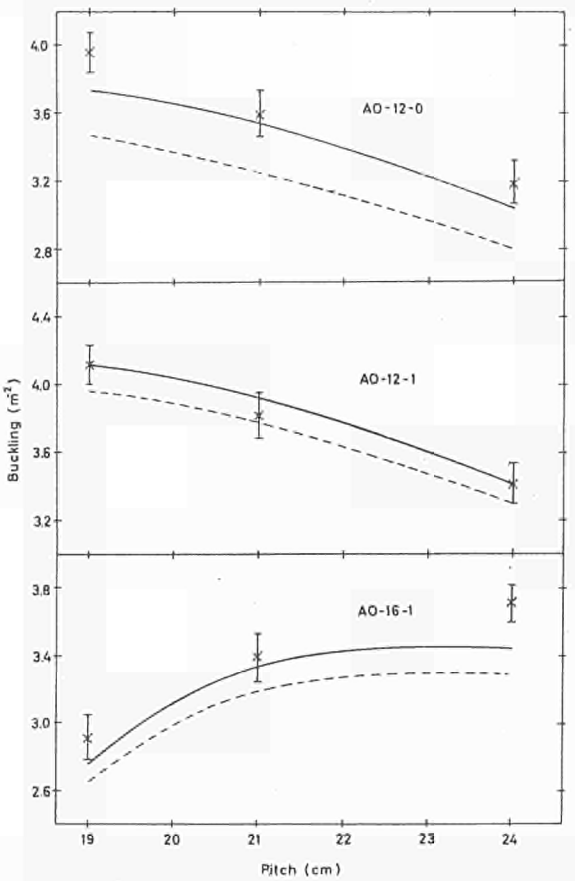

Fig. 5 - Comparison between measured and calculated values. Broken line: values calculated following the first correlation. Solid line: values calculated following the modified correlation.

buckling curves as a function of lattice pitch seems, on the contrary, to be reasonably respected.

A first correction was tried in the following manner. Some nuclear data entering into the lattice calculations are not well known. In particular, the curve of the scattering cross section of the hydrogen bound to the organic molecule is uncertain. The values used for the calculations were deduced from measurements made by Melkonian on light hydrocarbons (methane, ethane, etc.) [5]. The curve for butadiene was chosen among them since it represented the average value of the measurements. The maximum deviations from this curve of the other measurements carried out by Melkonian already attain 15% and there is reason to believe that the deviation is even greater for our type of organic liquid.

Moreover, the hypothesis of assuming a Maxwellian thermal spectrum at the heavy water temperature for averaging the hydrogen scattering cross section is somewhat arbitrary.

For the above-mentioned reasons we assumed as a first correction parameter the thermal scattering cross section of the organic liquid. In particular, it was found that by reducing the value previously used by about 30%, one obtains results which agree satisfactorily with the measured values (see fig. 5).

It seems therefore that, for the purpose of satisfying the immediate needs of the project and while still awaiting polyphenyl measurements, a « recipe » of such kind can be used.

It should be noted that such a criterion, even if it does make it possible to correct the neutron balance as a whole, nevertheless attributes every cause of the discrepancy between experimental and calculated values to the thermal utilization factor. This hypothesis is at least a matter of guesswork since, as previously mentioned, the values of A, a and b utilized in our calculations are those deduced from the French correlation for heavy water moderated and cooled lattices.

For all these reasons, we are now establishing a new correlation between experimental results and theory. This work is at present possible because we dispose of some other experimental buckling results from Canada and Denmark on organic cooled, heavy water moderated lattices. ■

7. NOTATION

K_∞	Infinite multiplication factor
L^2	Diffusion area
L^2_s	Slowing-down area
B^2_m	Material buckling
D_1	Fast diffusion coefficient
D	Thermal diffusion coefficient
β	Radial buckling
α	Axial buckling
γ	Constant taking into account axial flux asymmetry
f	Thermal utilization factor
ε	Fast fission factor
V_u	Volume of fuel in the cell
V_o	Volume of organic liquid in the cell
V_m	Volume of heavy water in the cell
$(\xi \Sigma_s)_o$	Slowing-down of the organic liquid
$(\xi \Sigma_s)_m$	Slowing-down power of the heavy water
P_B	Corrective factor taking into account the non-uniformity of the sources in calculating the resonance absorption

Acknowledgements. We wish to thank the « Département d'Etudes des Piles » of Saclay and especially Mr. Lourme for the continuous help given us while carrying out the experiments.

We have pleasure in expressing our special gratitude to Mr. Chabrillac and Mr. Ledanois for their assistance in processing the data collected and in interpreting the results.

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riassunto

ESPERIENZE CRITICHE SU RETICOLI DI OSSIDO DI URANIO NATURALE, MODERATI AD ACQUA PESANTE E REFRIGERATI MEDIANTE UN LIQUIDO ORGANICO

Una serie di misure è stata effettuata al Centro francese di Saclay da parte del Dipartimento di Fisica dei Reattori del CCR dell'Euratom.

Scopo di questi esperimenti era la determinazione del buckling materiale di nove diversi reticoli ORGEL formati con elementi di ossido di uranio naturale, moderati ad acqua pesante e refrigerati mediante un liquido organico. Questo rapporto descrive la tecnica sperimentale usata e indica i criteri per ottenere un soddisfacente accordo fra valori misurati e calcolati.

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