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EXPERIMENTAL INVESTIGATION OF THE U^{238} RESONANCE NEUTRON CAPTURE INTEGRAL OF URANIUM CARBIDE CLUSTERED FUEL ELEMENTS CONTAINING MODERATING COOLANTS

by

A. BOEUF and S. TASSAN

1966

ORGEL Program

Joint Nuclear Research Center
Ispra Establishment - Italy

Reactor Physics Department
Experimental Neutron Physics

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SUMMARY

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1. INTRODUCTION

The evolution of the heavy water moderated power reactors led to the consideration of lattices characterized by : large fuel cross sections per channel coupled with large lattice pitches, strong heterogeneities in the geometrical fuel configurations, cooling media different from the primary moderator. These features are particularly evident in the ORGEL type reactor under study at CCR Ispra, which is fuelled with clusters of few large-diameter rods (probably 4-rod or 7-rod clusters of uranium carbide), and cooled by an organic compound (1).

The calculation of the U^{238} resonance absorption in reactor lattices of this type presents considerable difficulties. As a consequence, the theoretical models developed for this calculation at CCR Ispra (2) involve various simplifying assumptions, whose validity needs experimental confirmation.

Resonance escape probability measurements provide the parameter expressing the U^{238} resonance capture in a lattice of fuel elements, i.e. the quantity directly useful for reactor calculations (3). The performance of measurements of the resonance escape probability requires however the assembly of a large number of full-size fuel elements equal to the element investigated, and the availability of a critical facility, or its equivalent, capable of generating a neutron flux of the order of 10^8 n/cm^2 sec at the center of the assembly. Such facility did not exist at CCR Ispra at the time when the experimental studies described in this report were undertaken.

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An alternate parameter expressing the U^{238} resonance neutron capture in reactor fuel elements is the "effective resonance integral", defined as "the lethargy integrated absorption cross section required to produce the same amount of absorption as actually takes place in the resonance, assuming that the flux has the value it would have in the absence of the resonance" (4). The energy interval of integration is usually taken from infinity to the energy corresponding to the effective cut-off of cadmium (about 0.5 eV, but depending on the specific experimental set-up).

The measurement of the effective resonance integral does not require a complex experimentation, but only a reduced-length segment of the fuel element investigated and a facility for inserting this sample at a fuel-vacant position, in the central zone of the lattice of almost any existing critical facility or low-power research reactor.

On the other hand, recent theoretical and experimental investigations (5,6) have shown that effective resonance integral experiments do not yield results which are immediately useful for reactor calculations, mainly due to the large deviation of the neutron spectrum at the surface of the fuel elements of the reactor lattice from the 1/E dependance which is required by the definition of the effective resonance integral. Consequently, the use of U^{238} effective resonance integral data for reactor calculations requires the evaluation of important correction terms, taking into account the actual variations of the neutron flux in space and energy.

Effective resonance integral experiments are still valuable to the reactor physicist, since they provide the information needed to test both the theoretical models and the differential data (cross section, etc.) used for the calculation of the neutron resonance capture in reactor fuel elements.

With the aim of providing this information, a series of measurements of the effective resonance integral for U^{238} capture of several U and UC fuel elements of various geometries was undertaken at CCR Ispra in 1963. At that time the only available experimental data on the effective resonance integral of clustered fuel elements were the old data (1957) by E.HELLSTRAND (16) and those by C.BERNARDER and K.JIRLOW (15), dealing with close-packed and loose UO_2 clusters, respectively. No consistent set of data existed for the effective resonance integral of UC, either in rod and in cluster form. In fact the situation has not significantly changed since then. The results of some of the experiments included in the above research project have appeared in previous publications (7,8).

This report describes the last phase of the project, i.e. the measurement of the U^{238} effective resonance integral of 4-rod and 7-rod uranium carbide clusters, of varying geometry and containing several cooling media; it also provides detailed information on the experimental techniques used, which was omitted in the previous publications due to lack of space.

2. MAIN FEATURES OF THE EXPERIMENTAL METHOD

Basically, the technique used consisted in comparing the U^{238} resonance neutron capture induced activities of the several UC clustered fuel elements considered to that of "reference" UC rods - having the same diameter as the rods of the clusters - whose effective resonance integrals were known with a good precision from a previous measurement (7).

The samples, contained in equal Cd-clad holders, were irradiated at the core center of the D_2O moderated ISPRA-I reactor. The neutron slowing down flux about the samples was monitored by suitable resonance detectors.

The features of this technique, with respect to the experimental conditions to be met for consistency with the standard definition of the effective resonance integral and the usual assumption of a spatially and lethargy-wise uniform neutron flux distribution used in theoretical calculations, are presented in some detail in ref. 7 and 9. For completeness, a part of the considerations made in ref. 7 are reported in the following paragraphs.

"According to the definition of effective resonance integral, the measurement of this quantity should be performed in a flux with a 1/E neutron slowing down distribution. In the last years, a departure from this ideal 1/E behaviour in actual reactor spectra has been predicted theoretically (5) and observed experimentally (6). The correction factor to be applied to the data, to take into account this departure which depends on the specific reactor configuration, is of difficult determination and represents at present the largest source of uncertainty in this kind of experiments (6,10,11).

The procedure of referring the resonance capture of the element investigated to that of a standard element, whose effective resonance integral in a 1/E slowing down neutron distribution is known, eliminates the need for the determination of such correction factor, provided the actual energy dependence of the neutron spectrum deviates only slightly from 1/E, and suitable experimental conditions are selected.

There are several reasons to consider that the former condition is met for the neutron spectrum in Ispra-I. In particular, measurements of the leakage neutron energy spectrum from a beam-port of the MIT CP-5 type reactor, which is very similar to Ispra-I, have shown that the epithermal component follows very closely the 1/E distribution (12) (Fig.1). Also, differential measurements by the fast chopper method, extended up to 10 keV, have put in evidence only a slight departure from the 1/E behaviour in the slowing down neutron distribution at the core center of the D_2O moderated reactor R-1 (13,14) - whose spectrum is not significantly different from the Ispra-I spectrum - corresponding to a few-percent correction to the measured U and UO_2 effective resonance integrals (6). That the Ispra-I spectrum does not depart considerably from the 1/E dependence was also indicated indirectly by the result of a measurement of the effective resonance integral of a U rod, normalized to the infinite dilution resonance integral of Au (see Appendix A.6.).

The deviations from the ideal behaviour in the flux at energies higher than 10 keV occurring in D_2O moderated reactors, have been shown not to have an appreciable effect on the measured U^{238} capture, at least for the S/M ranges usually investigated (11).

In such situation, the small spectral correction to the measured U^{238} resonance neutron capture induced activities can be assumed to be equal for the UC reference rods and the UC clusters over the whole S/M range considered in the present measurement. The error introduced into the results of the experiment by neglecting such correction, can be assumed to be trivial with respect to the other experimental errors."

The special features of the measurement, when performed on clustered fuel elements of practical interest - and particularly if these elements contain a hydrogeneous coolant - deserve some additional remarks.

A. Resonance flux monitoring

The normalization of the U^{238} resonance capture induced activities measured in successive irradiations requires monitoring of the resonance flux impinging on the sample. In the ideal case that the presence of the sample does not perturb the neutron flux (this case is approached when measuring "infinite dilution resonance integrals", using extremely thin samples), the choice of the detector type (i.e. energy sensitivity of the resonance detector) and location is relatively unimportant.

By definition, the measurement of the "effective resonance integral" requires the use of a sizeable sample, producing important depressions of the neutron flux at the energies of its main resonances. However, provided the sample is still relatively small, the neutron flux at the sample position can be considered not to be perturbed by the sample outside the resonance energies (more precisely outside the resonances "practical widths" (4)). In such case the resonance flux can be monitored at a position far from the sample, and the choice of the resonance detector is again immaterial. The above condition is generally met by "single-rod" samples, over the practical range of S/M values (i.e. 0.05 - 0.50 cm^2/g), as it has been put into evidence by several experimental tests (7,10).

On the contrary, the measurement of the effective resonance integral of a clustered fuel element requires the use of a massive sample, which may produce a considerable depression of the flux even outside the energies of its resonances (15). (It is reminded that the "length/diameter" value of the sample must be large enough for the sample to be representative of a full length fuel element, that is for the end-effects on the measured resonance capture to be negligible). In this case the resonance flux must be monitored as close as possible to the sample, or even inside the sample. However, when doing so, the resonances in the monitor may be shielded by the resonances of the sample and the monitor may fail to yield the right information.

The problem of correctly monitoring the resonance neutron flux impinging on the clustered fuel element sample is very delicate, and represents one of the largest sources of uncertainty for this kind of effective resonance integral measurements. Among the methods developed to overcome this problem one may mention a difference technique, using a double set of gold detectors (15,16). One of the gold package contained a thin foil and a thick one, the latter facing the cluster surface, the other package contained two thick foils with a thin one in between. The difference between the two thin foils induced activities was considered to be proportional to the resonance flux at the 4.9 eV resonance of gold, impinging on the cluster boundary. Another attempt to solve the problem consisted in using a monitor, with a low 1/v cross section and with its first activation resonance higher than the energy range of the sample resonances, so as to make the above shielding effect negligible.

The detector selected was Mo^{98}, on the basis that its first known activation resonance occurred at about 480 eV (10). The use of Mo^{98} allowed monitoring of the resonance flux around 480 eV, both at the cluster boundary and inside the rods of the cluster.

Although recent measurements (17,18) have shown the existance of a (n,γ) resonance at 12 eV in Mo^{98}, accounting for an appreciable part of the resonance integral of Mo (a discrepancy still exists with regard to the actual magnitude of such contribution) it is reasonable to assume that the 12 eV resonance of Mo^{98} is sufficiently lower than the next upper U^{238} resonance at 20 eV, not to be appreciably shielded by the latter, for a relatively large range of sample assemblies. This assumption is supported by published results of theoretical calculations, performed for a situation approximating the conditions typical of this kind of measurements (19), and by the evidence of experimental results (7,10).

For the present measurement Mo^{98} was chosen as the resonance flux detector, since the possibility of monitoring the flux inside the rods of the cluster was considered to be an important feature of the experimental technique, particularly in view of the comparison with the results of calculations performed using the "flat flux" approximation (see Section 4.2).

B. Neutron energy spectrum distortion

Another delicate aspect of the measurement of the effective resonance integral in fuel clusters containing a moderating coolant, is the distortion of the energy spectrum of the neutrons entering the cluster, due to collisions with the atoms of the cooling medium.

A satisfactory method for evaluating the effect of this distortion has not been so far developed. The main effects of such spectrum distortion are a modification of the resonance capture and an increase of the 1/v type capture in the fuel. The latter is caused by the neutrons entering the cluster with epi-cadmium energies (the incoming thermal neutrons are cut-off by the cadmium shield surrounding the sample) and slowed-down inside the cluster. The resulting sub-cadmium capture can be removed by surrounding with an additional cadmium sheet the rods of the clusters where the resonance detectors are placed; however this technique does not go without objections, since the presence of large amounts of cadmium inside the cluster may further distort the neutron energy spectrum, still leaving the need for the evaluation of a correction to take into account the increase in the epi-cadmium 1/v capture, which is obviously not affected by the cadmium shield around the rod.

The magnitude of both the neutron energy spectrum distortion and the 1/v capture excess is an increasing function of the slowing-down capability of the cooling medium. This may eventually lead to a limit for the validity of effective resonance integral measurements in clustered fuel elements, containing hydrogenated coolants and characterized by high coolant-to-fuel volume ratios, in the sense that it may result to be impossible to interpret the results of the measurements according to the standard definition of the effective resonance integral. Such clusters might be studied after improvement of the existing experimental methods, or through a modification of the definition of the effective resonance integral for clustered fuel elements, taking into account the above mentioned spectral distortion.

3. EXPERIMENTAL DETAILS

3.1. Experimental assemblies

The fuel elements investigated were 7-rod and 4-rod uranium carbide (UC) clusters, containing varying coolants at room temperature. The uranium carbide met the following specifications : U content : 95.28 ± 0.05% by weight; density : at least 99% of the theoretical density of UC (13.65 g/cm^3). The cooling media contained in the cluster were : air, 98.9% pure heavy water, "diphyl" (26.5% $C_{12}H_{10}$, 73.5% $C_{12}H_{10}O$), distilled light water.

The 7-rod cluster consisted of Al-clad UC rods, 25.2 mm in dia., arranged inside an Al pressure tube, 91 mm in dia. and 1.5 mm thick. A modified version of this fuel element contained six magnesium fillers, arranged against the pressure tube, which reduced the amount of coolant in the cluster. Details of the geometry of the 7-rod cluster are shown in Fig.2.

The 4-rod cluster consisted of Al-clad UC rods, 30.9 mm in dia., inserted inside a graphite matrix, having a diameter of 109 mm, in turn contained in a 2 mm thick Al tube. Around each fuel rod a 39.2 mm dia. thin Al tube generated a 2.5 mm thick annular void which was filled by the cooling medium. Details of the geometry of the 4-rod cluster are presented in Fig.3.

The samples designed for the experiment consisted of 25 cm long (7-rod cluster) and, respectively, 20 cm long (4-rod cluster) segments of the standard fuel elements.

Each rod of the cluster consisted of a pile of UC pellets, clad by a 1 mm thick Al tube; the lower end of the tube had a welded-in Al plug, the upper end was provided with a removable teflon plug (leak-proof). A 0.15 mm clearance existed between the UC pellets and the canning tube. The proper cluster geometry was obtained by means of Al end-plates (7-rod cluster) and by the holes pierced through the cylindrical graphite matrix (4-rod cluster).

The cluster-sample was kept inside a cylindrical Al container, having the same diameter as the outer tube of the standard cluster, which was closed by a leak-proof lid provided with a safety valve (to release the vapor pressure due to accidental overheating of the liquid coolant). The container was completely surrounded by a 1 mm thick sheet of cadmium, shielding the cluster sample against the thermal neutrons. This assembly was kept inside a second container (which was requested by reactor safety considerations) similar to the one above, but with its diameter matching the internal diameter of the 127 mm dia. reactor thimble used for the experiment . The 7-rod cluster set-up included a thin Al annular container, filled by heavy water, which eliminated the air gap existing between the sample and the internal surface of the thimble.

The cluster rods containing the UC detectors were wrapped, at their middle points, with a 10 cm high, 0.5 mm thick cadmium sleeve, in order to avoid the neutron capture in the rods due to the incoming epi-cadmium neutrons slowed down to sub-cadmium energies by the moderating coolant (and C matrix, in the case of the 4-rod cluster) contained in the cluster itself.

The cadmium sleeve was set in a recess of the aluminum cladding, in order not to modify the geometry of the cluster. In some 7-rod cluster irradiations such rods were kept bare, in order to put into evidence the magnitude of the neutron slowing-down in the cooling medium. The design of the 7-rod and 4-rod cluster assemblies, including details of several components, is shown in Fig.4 to 10.

3.2. Experimental facility

The irradiation of the samples was carried out in the Ispra-I reactor, a 5 MW, D_2O cooled and moderated, CP-5 type reactor at Euratom CCR Ispra, Italy (20). The reactor facility used was the 127 mm internal diameter aluminum thimble at the core axis. The experimental assembly was suspended to the stainless steel plug of the thimble by an aluminum rod, provided with two spherical joints. Such rigid supporting system assured a good reproducibility of the sample position with respect to the reactor core.

The length of the aluminum rod was adjusted so as to have the sample mid-plane at the reactor core mid-plane, i.e. at the position of minimum axial and radial flux gradient. This flux condition is essential for the correct performance of experiments of this kind, for the two following reasons :

A. The 1/E dependance of the neutron spectrum is closely approached in the central zone of the core of a finite-size critical reactor, while towards the core boundary leakage effects may cause significant distortions of the asymptotic neutron energy spectrum. As a consequence,

at the core periphery the neutron energy spectrum requirements set by the definition of the effective resonance integral may not be met, and experimental data obtained by irradiations performed in this zone may not be interpretable according to the above definition.

B. The correct normalization of the data obtained from irradiations of different samples requires a uniform flux (both in space and energy) in the zone of the sample.

The actual neutron energy spectrum in the U^{238} resonance region, existing at the experimental position in the Ispra-I reactor, could not be directly measured. However, several measurements performed in reactors whose neutron energy spectra are very similar to the Ispra-I spectrum have shown only minor departures from the ideal 1/E dependance of the flux and, in such condition, the method used for the present experiment does not require the determination of the neutron energy spectrum in the zone of the sample. More details about this aspect of the experiment are presented in Section 2.

The radial and axial distributions of the thermal and resonance flux inside the thimble at the sample position were measured by activation of Dy^{164} and Au^{197} detectors, respectively. The results of these preliminary measurements showed adequately flat flux distributions in the zone of interest; particularly important was the measured uniformity of the flux across the empty Cd-clad container of the cluster sample.

The sample irradiations in the thimble were performed at a reactor power of 1.2 KW, for 20 minutes. This power corresponds to a neutron flux around 10^{10} neutrons/cm^2 sec, as determined through a measurement of the induced activity of thin Au^{197} detectors by the standard beta-gamma coincidence technique (21). For the details and actual results of the above preliminary measurements one is referred to Appendix A.2.

The very large reactivity controlled by the Cd-clad samples, and the possibility for overheating the cooling media contained in the cluster samples (mainly due to gamma heating caused by the radiation emitted in the decay of the fission products resulting from the routine operation of the Ispra-I reactor at 5 MW) gave some concern with regard to the reactor safety requirements. As a consequence, pertinent calculations and preliminary tests on the equipment were performed, showing that the experiment was inherently safe if carried out with due consideration. This work is described in some detail in Appendix A.1.

3.3. Resonance detectors

The detectors for the U^{238} resonance capture in the fuel elements were 4 mm thick uranium carbide pellets, having the same diameter as the rods of the clusters, which were inserted at the mid-point of the representative rods in the cluster assemblies and, respectively, at the mid-point of the "reference" rods.

A high-precision technique (the electroerosion) was utilized for machining the thin pellets used as detectors, as well as the adjacent pellets in the UC rods, so as to assure a very good contact between detectors and rod pellets, and the absence of surface imperfections.

In fact, for the UC rod sizes considered, the U^{238} resonance capture is concentrated in a thin layer on the surface of the fuel rod (due to the depletion of the flux at the U^{238} resonance energies inside the UC rod) and therefore strongly depends on the aspect of the rod surface in the zone of the detector. An increase of the "effective surface" of the 4 mm thick UC detector due to :

1. imperfections of the lateral surface,
2. a misallignment of the detector with respect to the UC rod where it is inserted,
3. the existence of gaps between the flat surfaces of the detector and of the adjacent rod pellets through which the resonance neutrons stream inside the fuel rod,

may produce an important addition to the neutron resonance capture of the detector, i.e. in the measured quantity.

For the actual experimental assemblies, the three effects indicated above were investigated separately, by standard methods; the only significant addition to the measured resonance capture was found to be the 0.3% increase produced by the maximum possible misallignment between detector and fuel rod (0.15 mm, namely the clearance existing between the rod and the aluminum cladding tube).

3.4. Measurement of the U^{238} neutron capture induced activity

The radiative neutron capture by U^{238} produces the well known decay chain

$$U^{238}\ (n,\gamma)\ U^{239} \xrightarrow[\beta^-,\gamma]{23.5\ \text{min}} N_p^{239} \xrightarrow[\beta^-,\gamma]{2.33\text{d}} Pu^{239} \qquad (1)$$

The detection of either the U^{239} or the Np^{239} induced activity can be used as a measure of the neutron capture by U^{238}.

The technique employed in the present experiment was based on the measurement of the intensity of the 103 keV photopeak from the Np^{239} decay (actually a 106 keV gamma ray and a 103 keV internal conversion X-ray from the deexcitation of Pu^{239} (22)), using a 50 keV window centered on the peak so as to give a 0.5% variation in the count rate per 1 Volt bias drift.

Three types of spurious activities - i.e. not caused by radiative neutron capture in U^{238} - contaminate the activity measured in the energy range considered, i.e. : room background, U natural activity, activity from the fission products decay. The first two activities were easily corrected for by the usual procedure (e.g. for the natural activity, by counting the activity of a non irradiated UC detector and normalizing it to that relative to the actual detector by the proper weight ratio). The error introduced by applying these two corrections to the data was definitively negligible.

The fission product activity in the energy region of interest was corrected for experimentally, by a technique which is described in some detail in Appendix A.5. The application of such correction, whose magnitude was in the order of 2% of the measured Np^{239} activity, increased the margin of error of the data by about 0.2%.

Since uranium exhibits a strong self-absorption for the gamma rays emitted in the 100 keV range (23), the direct counting of the radiation emitted by the 4 mm thick UC detectors was not feasible. It was then necessary to count the activated detectors in the form of liquid solutions, characterized by a much lower U atom density, for which the gamma ray self-absorption correction factor could be easily evaluated.

To this effect, the UC detectors recovered from the irradiated fuel rods were calcinated at 1000°C and the resulting U_2O_3 powder was dissolved in a hot HNO_3 solution. From such solution 3 cc samples were transferred into cylindrical teflon capsules, precision machined for good-geometry gamma counting. Several successive weight and volume determinationsassured the precise knowledge of the amount of U contained in each sample. The details of the procedure followed for the preparation on the liquid samples for counting, which represented one of the most delicate aspects of the experimental technique, are described in Appendix A.3.

The homogeneization resulting from the dissolution process also eliminated the possible effect of the strongly non-uniform activation of the large-diameter UC detectors on the geometric efficiency of the NaI crystal-single channel analyzer counting units used. In fact, as indicated previously, the U^{238} resonance absorption is concentrated on the outer layer of the fuel rods, but in a different way for the cluster rods and the isolated reference rod: this may result in a differential effect on the geometric efficiency of the NaI crystal, which cannot be taken into account in simple way (24).

A phenomenon often occuring when counting high disintegration rates, is the count rate dependent drift in the height of the photomultiplier output pulses. In order to minimize the effect of this phenomenon , which cannot be easily corrected for, close counting rates from all 3 cc samples were obtained by a suitable selection of the U concentration in the solutions from which the samples were prepared.

The variation of the U atom density in the samples for counting (from 0.49 to 0.51 gr/3 cm^3 for the 7-rod UC cluster, and from 0.735 to 0.760 gr/3 cm^3 for the 4-rod UC cluster) required the use of a correction factor, to account for the corresponding variation in the self-absorption of the emitted 103 keV gamma rays. This correction factor, which was determined experimentally by counting the activities of 3 cc liquid samples of varying U concentration, ranged from 0.91 to 0.906. The application of the correction factor for the gamma ray self-absorption introduced in the data a less than 0.1% uncertainty. The technique used for the determination of the gamma ray self-absorption in the counting samples, as well as the results obtained, are outlined in Appendix A.4.

3.5. Counting equipment

The equipment used for measuring the U^{238} neutron capture induced activities of the 3 cc samples consisted of two 2x2 in. NaI(Tl) crystal-single channel analyzer units, the crystals being placed at 180° on either side of the sample inside a 5 cm thick lead shield. Such arrangement of the crystals made the combined efficiency of the counters practically independent of the minor variations which could be expected in the sample-to-crystal spacings (since the dependence of counter efficiency on small variations of sample-to-crystal spacing is linear in the first approximation

the effect on one counter is balanced by the equal - but opposite in sign - effect on the other counter).

The dead time of the counting equipment was determined by the two-source technique (25), using two equal 3 cc liquid sqmples, with induced activities in the range of the activities measured in the actual experiment, and counter settings at the values selected for the experiment. By this procedure one obtained the value for the dead time of the equipment corresponding to the actual experimental conditions, i.e. taking into account the effect of count-rate, spectral composition of measured radiation, selected energy band, and equipment settings.

The equipment over-all dead time thus determined was 4 μsec, and constant over the observed count-rate interval from 50.000 to 300.000 cpm.

3.6. Resonance flux monitoring

3.6.1. Flux at the sample position

Two techniques were used for normalizing the U^{238} resonance neutron capture to the resonance flux at the experimental assembly. They consisted in monitoring :

A. The resonance flux impinging on the cluster boundary and, respectively, on the surface of the reference UC rod (boundary resonance flux monitoring technique) ;

B. The average resonance flux across the representative rods of the cluster and, respectively, across the reference UC rod (average resonance flux monitoring technique).

In the first case, the Cd-clad Mo detectors, 0.10 mm thick and 4 mm in diameter, were placed at the sample mid-plane, on the surface of the cluster outer-ring rods next to the Cd-clad pressure tube, and on the surface of the reference UC rod. In the second case, the Mo detectors, 0.10 mm thick and 30.9 mm or 25.2 mm in diameter, were inserted inside representative rods of the cluster and inside the reference UC rod.

The neutron capture induced activities of the Mo^{98} monitors were determined by measuring the intensity of the photopeak of the 140 keV gamma ray resulting from the 67 hr half-life decay of Mo^{99}. More exactly the decay followed was the equilibrium state of the complex decay of the 6.2 hr half-life Tc^{99m} and the 67 hr half-life Mo^{99} (22). The corresponding decay scheme is reported for convenience in Fig.11.

The correction factor for the self-absorption of the 140 keV gamma ray emitted by the 0.1 mm thick Mo detectors was calculated by the standard formula (23)

$$T = \frac{1}{\lambda t}\left(1 - e^{-\lambda t}\right) \qquad (2)$$

where :

- t is the detector thickness, in cm ;
- λ is the linear coefficient for the gamma absorption at the measured energy, in cm^{-1} ($\lambda = 4.66\ cm^{-1}$ for Mo at a gamma energy of 140 keV) ;
- T is a transmission factor expressing the fraction of the total radiation resulting from the disintegration rate of the detector which is emitted from the surface of the detector.

The magnitude of the transmission, as calculated by Eq.2, was in the order of 0.98; however the relative correction factor, corresponding to the observed variations in the thickness of the Mo detectors, was 1.00 ± 0.002, and the uncertainty introduced in the data by applying such correction was definitively negligible.

The induced activity of the Mo detectors was measured by the same NaI(Tl) crystal-single channel analyzer counting equipment utilized for the detection of the U^{238} neutron capture induced activities of the 3 cc liquid samples. Since the highest measured Mo^{98} activities were in the order of 50.000 cpm, only a very minor correction was required for the dead-time of the counting equipment and the accurate determination of the dead-time, corresponding to the measured radiation, was not necessary. For this reason, the count rates from the Mo detectors were corrected for using the 4 usec dead-time measured for the U^{238} neutron capture induced radiation. The margin of error of the data was not affected by this approximation.

3.6.2. Unperturbed flux

For the purpose of providing an experimental indication of the magnitude of the depression of the slowing-down neutron flux caused by the large clustered fuel element samples even outside the U^{238} resonance energy range, during each sample irradiation the "unperturbed" flux was monitored by activation of Au detectors.

The detectors (12 mm dia., 0.2 mm thick pure Au disks) were contained inside 0.8 mm thick cadmium boxes and fixed in a recess on the Al rod supporting the sample holder, at a distance of 20 cm from the upper end of the UC sample. This distance was determined so as to insure a negligible shielding of the 4.9 eV resonance of the Au monitor by the lowest U^{238} resonance of the UC sample (see also Section 2 and Appendix A.6.1.).

Due to the almost complete self-shielding of the 4.9 eV Au resonance inside the outer layers of the 0.2 mm thick foils, the activation of the gold monitors was independent of the foil thickness, for the thicknesses considered. The Au induced activities were measured by following the 410 keV, 2.7 d half-life gamma activity from the decay of Au^{198}.

4. EXPERIMENTAL RESULTS

4.1. Analysis of data

The U^{238} neutron resonance capture in the UC clusters relative to the corresponding capture in the reference UC rods, R_i, was obtained from the following relationships, valid respectively when one monitors the resonance neutron flux at the cluster boundary (R_1), and the average flux across each representative rod of the cluster (R_2).

For the 7-rod cluster :

$$R_1 = \frac{1/7\,(U_0 + 6\,U_1)}{U_r}\ \frac{M_r}{M_c} \qquad (3)$$

$$R_2 = \frac{1/7\ (U_o/\bar{M}_o + 6\ U_1/\bar{M}_1)}{(U_r/\bar{M}_r)} \qquad (4)$$

For the 4-rod cluster :

$$R_1 = \frac{U/M_c}{U/M_r} \qquad (5)$$

$$R_2 = \frac{U/\bar{M}}{U/\bar{M}_r} \qquad (6)$$

In the above equations :

U, U_o, U_1, U_r are the measured U^{238} neutron capture induced activities of the UC detectors irradiated respectively in a rod of the 4-rod cluster, in the center rod and in the outer-ring rod of the 7-rod cluster, and in the reference rod.

M_c, M_r are the measured activities of the Mo foils placed at the boundary of the cluster and at the surface of the reference rod, respectively.

$\bar{M}$, $\bar{M}_o$, $\bar{M}_1$, $\bar{M}_r$ are the measured activities of the Mo foils placed respectively inside a rod of the 4-rod cluster, inside the center rod and the outer ring of the 7-rod cluster, and inside the reference rod.

The effective resonance integral of the cluster, RI_i $(i=1,2)$ is expressed in function of the measured quantity R_i, by the equation

$$R_i = \frac{k_c RI_i + \sigma_c^{1/w}}{k_r RI_r + \sigma_r^{1/w}} \qquad (i = 1,2) \qquad (7)$$

where :

RI_r is the effective resonance integral of the reference UC rod in a neutron flux with the 1/E behaviour ;

$\sigma_c^{1/v}, \sigma_r^{1/v}$ are the contributions (in barns) of the 1/v varying part of the cross section to the U^{238} neutron capture above the energy corresponding to the effective cut-off of the cadmium shield, in the cluster and in the reference rod, respectively ;

k_c, k_r are correction factors taking into account the departure of the actual neutron flux from the ideal 1/E dependence required by the definition of effective resonance integral ;

the subscripts (1,2) refer to the values of RI_i obtained using the values of R_1 and, respectively, R_2.

The magnitude of the correction factor k_c, k_r depends on the unperturbed resonance neutron spectrum of the environment where the experimental assembly is placed and, to a minor extent, on spectral distortions produced by the experimental assembly itself. Such distortions in the resonance neutron spectrum are in turn dependent on the resonance structure of the fuel material (U, Th, etc.), since the neutron flux is depleted at the energies of the most important resonances, and on the mass and geometrical dimensions of the experimental assembly (which may include coolants acting as neutron moderators, as in the present case). Then, in general, the correction factor k_c, to be applied to the measured neutron resonance capture in the cluster assembly, differs from the corresponding correction factor, k_r, for capture in the reference single-rod assembly.

Under the condition that the actual neutron energy spectrum does not deviate seriously from the 1/E behaviour, i.e. that the correction factor k departs only by a few percent from unity, and that the sample and the reference standard have the same resonance structure, it is reasonable to assume $k_c = k_r = k$. Furthermore, when RI is much larger than $\sigma^{1/v}$ for both the sample and the standard, and RI_c is relatively close to RI_r, the ratio represented by Eq.7 is not appreciably altered if the correction factor k is neglected. In the present case the above requirements are met, being :

$$k \sim 1.05, \quad RI_{c,r} \gtrsim 10\,\sigma^{1/v}, \quad 0.8 < RI_c/RI_r < 1.0.$$

The 1/v contribution to the U^{238} neutron capture, is slightly larger in the clustered fuel element than in the reference single-rod, mainly due to the slowing down of neutrons inside the cadmium-clad Al pressure tube of the cluster by the moderating coolant, which increases the neutron density at the lower energies (the activation of a 1/v absorber is mostly caused by capture in the low energy range). The difference between $\sigma_c^{1/v}$ and $\sigma_r^{1/v}$ is however not important (a calculated value $\sigma_c^{1/v}/\sigma_r^{1/v} = 1.05$ has been reported for UO_2 clusters containing a hydrogeneous coolant, and with a coolant-to-fuel volume ratio higher by a factor of three than that characterizing the clusters investigated in this experiment (15)) and has been neglected in the present case, where $\sigma^{1/v}$ represents only about one tenth of the measured resonance integrals. The value of $\sigma^{1/v} = 1.2$ barns has been calculated on the basis of the thickness and geometry of the cadmium shield surrounding the experimental assembly (25).

With the above assumptions Eq.7 is reduced to the following simpler expression

$$R_i = \frac{RI_i + 6^{1/N}}{RI_r + 6^{1/N}} \qquad (8)$$

The error implied in the use of Eq.8, instead of the more exact expression given by Eq.7, was considered to be unimportant with respect to the random experimental errors affecting the measured values of R_i.

The values for the effective resonance integrals, RI_i, of the clusters studied were inferred from Eq.8, using the value of RI_r calculated by the empirical expression for the effective resonance integral of UC, RI_{UC}, determined by A.BOEUF and S.TASSAN (7) :

$$RI_{UC}\ (\text{barns}) = 3.14 + 26.95\sqrt{S/M} \qquad (9)$$
$$0.09 < S/M\ (cm^2/g) < 0.30$$

The curve represented by Eq.9 is reported for convenience in Fig.12. From this curve the values of the effective resonance integrals of the UC reference rods, with diameters 30.9 mm (S/M = 0.094 cm^2/g) and 25.2 mm (S/M = 0.116 cm^2/g) are 11.4 barns and 12.4 barns, respectively.

4.2. R e s u l t s

The results of the experiment are reported in Tables 1 to 5. The measured values of R_1 (boundary resonance flux monitoring) and R_2 (average resonance flux monitoring) are listed in Table 1 (4-rod cluster data) and Table 2 (7-rod cluster data). The quoted errors are the standard deviations of the mean of at least 4 independent measurements.

They include, besides the contribution of the counting statistics uncertainty, the effect of the random errors deriving from variations in UC density and composition, imperfections in the UC detector geometry, misallignment of the detector with respect to the UC rod, UC dissolving procedure, correction for the self-shielding of the gamma radiation emitted by the detector, counting equipment drifts, fission product correction, etc.

Since the remaining sources of error, which were systematic to the measurements, were estimated to be unimportant with respect to the sources of random errors, it was concluded that the uncertainty inferred from the spread in the results was a reasonable evaluation of the actual overall error involved in the experiment. This error was typically $\pm$2%.

Also listed in Tables 1 and 2 are the values of RI_1 and RI_2 inferred from the measured R_i's by means of Eq.8. Including the $\pm$4.5% uncertainty quoted for the values of RI_r when inferred from Eq.9, the total error estimate for the values of $RI_{1,2}$ is $\pm$5%.

The values of RI_i relative to the center rod and the outer rods of the 7-rod cluster are reported in Table 3. The values of R_i and RI_i, as inferred by means of Eq.3,4 and 8 from the results of the measurements performed with the cluster rods containing the resonance detector not clad by cadmium, are listed in Table 4. The RI_i's thus obtained are not effective resonance integrals according to the standard definition of this parameter, since they include the contribution of the capture of neutrons slowed down to sub-cadmium energies by collisions with the moderating coolant contained inside the cluster. These data are presented in order to put into evidence the importance of such neutron slowing down process.

In the last columns of Tables 1 and 2 are listed the values of the relative resonance integral of the clusters, R, as calculated by C.CASINI and W.DE HAAN (2) by means of the NORDHEIM method (26), using the "flat flux" approximation and a DANCOFF correction factor obtained from the code SHOCK, under the assumption that only the collisions with the light nuclei of the moderating coolant (H,D) remove the neutrons from the original energy band.

For more details about the calculation one is referred to the original publication. It is noted that, for reasons of consistency between theoretical and experimental conditions, the comparison with the measured data should be made using the values of R_2 (i.e. obtained using the "average resonance flux monitoring" technique); in fact the procedure of dividing the resonance neutron capture induced activity of each rod in the cluster by the average Mo activity in the same rod - which is equivalent to normalize the U^{238} resonance capture to a uniform resonance flux across the cluster - corresponds to the "flat flux" approximation used for the calculation.

The values of R_1 and RI_1, as obtained from Eq.3 and 8 by normalizing the U^{238} neutron capture activities to the "unperturbed" resonance flux monitored by the Au detectors, are listed in Table 5. These data are presented to give evidence of the considerable local flux depressions caused by the cluster samples containing hydrogeneous cooling media.

The RI's determined by normalization to the unperturbed resonance flux could be used for comparison with the results of theoretical calculations of the effective resonance integral, provided the calculation would provide also the magnitude of the local flux depression produced by the sample itself. However, such calculation would be very complex and it is dubious whether it could be carried out with meaningful accuracy.

5. DISCUSSION AND CONCLUSION

The analysis of the data listed in Tables 1 to 5 leads to the following considerations :

A. The measured effective resonance integrals of the 7-rod UC cluster do not show an evident dependence on the type of coolant contained in the cluster (lines 3, 4 and 5 of Table 2). This behaviour may be explained qualitatively on the basis of the open geometry of the 7-rod cluster, implying that only a minor fraction of the coolant contained inside the cluster pressure tube affects the resonance capture in the fuel (in fact as shown in Fig.2, most of the cooling medium is concentrated in the six triangle-shaped areas outside the boundary of the cluster).

B. As expected, the presence of the organic coolant and of the graphite matrix affects to some extent and, respectively, to a negligible extent the resonance capture in the fuel of the 4-rod cluster investigated (Table 1).

C. The fraction of the neutrons entering the 7-rod cluster boundary with epi-cadmium energies and slowed down inside the cluster, due to collisions with the cooling medium, is an increasing function of the moderating power of the coolant. This behaviour is put into evidence by the comparison of the results obtained from the irradiations carried out with the rods containing the detectors bare and, respectively, wrapped with cadmium (Tables 1, 2 and 4). The increase in the capture in the rods, due to such 1/v-type capture contribution in the thermal region, over the true resonance capture, varied from about 6% in the case of the D_2O coolant, to about 30% in the case of the H_2O coolant. It follows that the correct performance of resonance integral measurements for the cluster types considered, requires indeed the use of a double cadmium shield (around the whole cluster and each measuring rod, respectively), or of equivalent techniques, for taking into account this important neutron slowing-down effect.

D. The values for the effective resonance integral obtained by monitoring the average resonance flux across the representative rods of the cluster are systematically higher than the corresponding values determined by monitoring the resonance flux impinging on the cluster boundary (compare columns 3 and 5 in Table 1 and 2). Such behaviour is the result of the measured depression of the Mo^{98} activation towards the center of the cluster; this depression increases with the improvement of the slowing down capability of the cooling medium and may be therefore associated, in part with the local flux depletion caused by the large cluster sample, and in part with the effect of the distortion of the neutron energy spectrum inside the cluster on the capture by the 12 eV resonance of Mo^{98}.

The quantitative separation of the two effects by calculation is difficult, also due to the discrepancies existing among the measured parameters for the 12 eV resonance of Mo^{98}. For more details on the processes mentioned in the above paragraph one is referred to Section 2 in the text.

E. The values for the effective resonance integral obtained by monitoring the local resonance flux are systematically higher than the corresponding values measured by monitoring the unperturbed resonance flux (compare Tables 1, 2 with Table 5). The difference is a consequence of the local flux depression produced by the large cluster samples. Since for the 7-rod cluster this difference increases considerably with the improvement of the slowing down capability of the cooling medium, it seems that the above flux depression is caused mainly by neutron moderation and absorption by the coolant contained in the Cd-clad sample holder.

F. The comparison between experimental and calculated values of the relative resonance integral, R shows an agreement which can be deemed satisfactory, when considering the magnitude of the experimental errors and the simplifying assumptions implied in the calculation (compare columns 4 and 6 in Tables 1 and 2).
This agreement seems to indicate that the experimental techniques and calculation methods used are essentially valid (even if it can be in part due to compensation of errors) and represents a basis for further experimental and theoretical activity in this field. In this view, the program for the next period includes :

- A systematically study of the approximation involved in the NORDHEIM method for resonance absorption calculation, using a newly developed multi-group, multi-region method for calculating fluxes and resonance absorption in a heterogeneous lattice cell (28). In this method the transport equation is solved by the collision probability technique, the collision probabilities being calculated as suggested by R.BONALUMI (29).

- A series of measurements of U^{238} resonance absorption (initial conversion ratio) in full lattices of organic cooled UC clustered fuel elements, D_2O moderated, using the E.C.O. (Essais Critiques Orgel) critical facility now in operation (30).

APPENDIX

A.1. CONSIDERATIONS REGARDING THE SAFETY OF THE EXPERIMENT

A.1.1. General

The main problems to be considered from the point of view of the safety of the experiment were :

A. The effect of the Cd-clad sample, placed inside the central thimble of the Ispra-I reactor at the core mid-plane, on the reactivity of the reactor ;

B. The increase in the temperature of the sample - by fission and gamma heating - during irradiation in the above position.

The general phylosophy of this analysis was to use simple calculation models coupled with conservative assumptions regarding flux values, heat dissipation, etc., and to resort to direct experimental tests when the simplified calculation failed to provide the required safety margin.

The following considerations are relative to the 19-rod U cluster, which was the first clustered fuel element irradiated in the facility. Similar considerations would apply to the UC clusters.

A.1.2. Reactivity worth of the experimental assembly

The cluster and single-rod samples, being contained inside the same Cd-clad holder, were essentially equivalent to cylindrical body black to thermal neutrons, having a diameter of about 8 cm and a height of about 34 cm. Such black body, placed at the center of a core with a volume of about 3×10^5 cm^3, had a negative reactivity worth of several thousands p.c.m.

The hydrogeneous cooling medium, contained in some cluster samples, slowed down the epi-cadmium neutrons crossing the Cd-shield, so that a large fraction of them was captured by the shield on their way out; this effect increased to some extent the negative reactivity worth of the cluster samples filled with moderating coolants.

The reactivity worth of the individual samples was therefore determined experimentally by subcritical approach. The values thus obtained lied in the neighborhood of 6000 pcm.

Since the reactivity worth of the sample placed in the central thimble was a rapidly varying function of its position with respect to core center (with its maximum at the core center), several precautions were taken in order to minimize the chances for either a variation of the sample position in successive measurements and a accidental fall of the sample to the bottom of the thimble.

Thus the sample was supported by a rigid aluminum rod fixed to the stainless steel plug of the thimble(the position of the plug with respect to the reactor top plate was accurately reproducible), two additional safety chains connected the sample to the plug, and a shock-absorbing platform was located at the thimble bottom reaching few millimeters below the sample lower end. The shock-absorber was designed so as to resist to the impact of the sample falling from a height corresponding to the reactor top plate level.

A.1.3. Increase in the sample temperature during irradiation

The increase in the temperature of the sample located in the central thimble of the Ispra-I reactor was essentially caused by the degradation of the energy liberated by the fission occurring in the sample irradiated at a reactor power of 1.2 kW, and by heating from the absorption of the gamma rays emitted in the decay of the fission products, built-up in the core during the routine operation of the reactor at its nominal power (5 MW).

The heat released by fission in the sample exposed to a flux around 10^{10} neutrons/cm^2 sec (i.e. the flux corresponding to the 1.2 kW power at which the sample irradiations were performed) was calculated by simple formulas to cause a negligible temperature increase, even under the most unfavorable assumption of an adiabatic process. The outline of the simplified calculation carried out for the determination of the fission product decay gamma heating of the sample, is reported in the following paragraphs.

A. Outline of the calculation of the fission product decay gamma heating

The Ispra-I core was approximated by a cylindrical core having the following dimensions :

$$H_{core} = 60 \text{ cm}$$
$$\emptyset_{core} = 84 \text{ cm}$$
$$V_{core} = 3.32 \times 10^5 \text{ cm}^3$$

The volume fractions of the core components were assumed to be :

for uranium : $f_u = 0.00141$

for aluminum : $f_{al} = 0.054$

for heavy water : $f_w = 0.9446$

The core power density corresponding to the maximum power (5 MW)

$$P = \frac{5 \times 10^6}{3.32 \times 10^5} = 15.1 \text{ watt/cm}^3$$

was considered uniformily distributed throughout the core.

From ROCKWELL'S "Reactor Shielding Manual", page 35 (31), the rate of delayed gamma energy release, two days after shut-down of a reactor operated at 1 Watt for infinite time is :

Table 1

group of delayed gammas	effective energy in MeV/photon	rate of energy release in MeV/sec x watt
I	0.4	1.1×10^9
II	0.8	6.5×10^9
III	1.3	0.023×10^9
IV	1.7	3.2×10^9
V	2.2	0.056×10^9
VI	2.5	0.26×10^9

Multiplying the values of the last column by P(watt/cm^3) the delayed gamma volume sources in the Ispra-I were obtained as :

$$S_1 = 16.6 \times 10^9 \text{ MeV/cm}^3 \times \text{sec}$$

$$S_2 = 98 \times 10^9$$

$$S_3 = 0.35 \times 10^9$$

$$S_4 = 48.3 \times 10^9$$

$$S_5 = 0.845 \times 10^9$$

$$S_6 = 3.93 \times 10^9$$

Making use of the core volume fractions, the linear energy absorption coefficients of the core were calculate for each of the energy groups listed in Table 1. Since these coefficients differred only slightly from each other, an average value was used for all the six energy groups, namely

$$\lambda_a \text{ (core)} = 0.035 \text{ cm}^{-1}$$

Due to the high volume fraction of the heavy water in the core, this coefficient was very close to the value for pure D_2O.

The sample, so far as the heat generation by gamma absorption was concerned, was considered to be confined to the inner container only, where the uranium was located. The dimensions of this "absorbing" part were :

$$H_{sample} = 34 \text{ cm}$$

$$\phi_{sample} = 8 \text{ cm}$$

$$V_{sample} = 1708 \text{ cm}^3$$

The volume fractions of its components were :

$$\text{for uranium} = f'_u = 0.377$$

$$\text{for aluminum} = f'_{al} = 0.190$$

$$\text{for organic} = f_{og} = 0.308$$

the total weight was 13.683 g; the thermal capacity was 3152 joule/°C. Moreover, making use of the volume fractions listed above, the linear energy absorption coefficients of the sample were calculated for each of the energy groups mentioned in Table 1.

$$\mu_1 = 1.51 \text{ cm}^{-1}$$

$$\mu_2 = 0.47$$

$$\mu_3 = 0.31$$

$$\mu_4 = \mu_5 = \mu_6 = 0.26$$

From ROCKWELL's book, page 365, the gamma flux at the middle point of the axis of a cylinder, having a gamma volume source S_v and a linear energy absorption coefficient μ_a; is given by $\phi = 0.79\ S_v/\mu_a$, having considered a build-up factor B=1.

Making use of this equation, the numerical values of the source $S_1 \ldots S_6$ and μ_a, were obtained

$$\phi_1 = 3.747 \times 10^{11} \text{ MeV/cm}^2 \times \text{sec}$$

$$\phi_2 = 22.120 \times 10^{11}$$

$$\phi_3 = 0.079 \times 10^{11}$$

$$\phi_4 = 11.000 \times 10^{11}$$

$$\phi_5 = 0.191 \times 10^{11}$$

$$\phi_6 = 0.887 \times 10^{11}$$

The rate of energy release in the sample per cm^3 was then

$$W = \sum_{j=i}^{6} \phi_j \ (MeV/cm^2 \times sec) \times \mu_j^{(sample)} \ (cm^{-1}) =$$

$$= 19.22 \times 10^{11} \ (MeV/cm^3 \times sec) \ ;$$

the rate of energy release in the whole sample was $W.V_{sample} = 525$ watt; the rate of energy release in the sample per gram was

$$\frac{W.V_{sample}}{\text{weight of sample}} = 38 \ MWatt/g$$

The evaluation of the increase in the sample temperature, corresponding to the estimated energy absorption rate in the sample during irradiation, is strongly dependent on the assumptions made about the mechanism of heat transfer from the sample to the D_2O moderator. The most conservative assumption is to consider the sample perfectly insulated. In this case the sample temperature increases linearly with time, with a slope which depends on the thermal capacity of the sample.

Assuming that the energy released by gamma absorption in the sample was completely absorbed in it (adiabatic case), the rate of temperature increase when the outer container was empty (thermal capacity of the sample equal to 3152 joule/°C) resulted :

$$\frac{\Delta T}{sec} = \frac{W.V_{sample}}{\text{thermal capacity of sample}} = 0.17 \ °C/sec$$

When the outer container was filled with heavy water, the thermal capacity of the sample became 10.868 joule/°C; in this case the rate of temperature increase (in the adiabatic assumption) became

$$\frac{\Delta T'}{sec} = \frac{W.V_{sample}}{10.868 \text{ joule } °C^{-1}} = 0.048 \text{ °C/sec}$$

After 30 minutes of irradiation of the sample the temperature increase was than 306°C in the first case, and 86°C in the second case.

Having verified that the assumption of adiabatic process led to an excessive temperature rise in the sample (provided the estimate of the gamma heating was correct), a more realistic assumption was made, taking into account the heat dissipation by convection to the air in the thimble and by conduction to the thimble walls and D_2O moderator. Again two limiting cases were considered (32) :

a. heat dissipated only by free convection to the air in the thimble ;
b. heat transmitted only by conduction to the thimble wall.

In case a. the temperature rise in the sample with a heat capacity around 11.000 joule/°C was evaluated to be 80°C after a 30 minutes exposure. In case b. it was calculated that direct contact between sample wall and thimble wall on 2% of the sample lateral surface resulted in a temperature rise of about 10°C at steady state.

It was concluded that, by improving the contact between sample and thimble walls, the heat transfer to the thimble would have been adequate to limit the temperature rise in the sample to some tens degrees during the 30-60 minutes irradiation time.

B. Measurement of the temperature rise in the sample

Due to the importance of a correct evaluation of the sample temperature increase during irradiation, with respect to both the reactor safety and the experimental requirements (the effective resonance integral is a function of the sample temperature, as a consequence of the Doppler broadening of the resonances) the above analysis was completed by direct measurements of the temperature in the actual sample.

The measurements were performed by means of two thermocouples, placed inside the organic moderator contained in the cluster and, respectively, inside the heavy water jacket filling the space between the cluster and the thimble wall. Each thermocouple was of the Ni-NiCr type; the two wires were surrounded by a metallic cladding which was soldered to the wires at the hot point. The Ni-NiCr wires were led to an electrical junction box fixed on the Al supporting rod of the sample, from there two extension wires reached the measuring instrument consisting of a high-precision resistance bridge. Both thermocouples were tested, prior to use, at three different temperatures.

The increase in the sample temperature due to gamma heating and to fission heating was determined separately, by exposing the sample, placed inside the central thimble of Ispra-I at the core mid-plane, to the radiation field existing when the reactor was shut-down and, respectively, operated at 250 W power. The results of the two measurements are presented in Fig.13 and 14, showing the curves of the temperature increase vs. exposure time.

The observed increase in the temperature of the organic compound (diphyl) in the cluster, due to gamma heating only, was about 20°C after one hour exposure; irradiating the sample at about 250 W for 20 minutes caused an additional temperature rise of 2°C.

By extrapolation of these data to a 20 min. irradiation of the sample at the reactor power of 1.2 kW used for the measurements, one obtained an overall temperature increase of the organic liquid of about 30°C, after one hour exposure. Such temperature rise was well below the limits set for the reactor safety (the boiling point of diphyl is about 250°C), as well as by the experimental requirement of negligible Doppler effect. The observed rise in temperature of the D_2O contained in the sample assembly was unimportant from the safety point of view.

In order to eliminate any chance of break-down of the sample, due to the vapor pressure generated by accidental overheating of the liquids contained in it (such overheating could have occurred only in the extreme conditions resulting from a sequence of wrong operations), the three containers of the sample were provided with pressure safety valves. The tightness and break-down pressure of these valves were periodically tested.

A.2. DETERMINATION OF THE ABSOLUTE FLUX AND OF THE RELATIVE FLUX DISTRIBUTION INSIDE THE ISPRA-I CENTRAL THIMBLE

A.2.1. Relative flux distributions

The determination of the spatial distribution of the neutron flux inside the 127 mm dia. thimble at the core axis of the Ispra-I reactor, was performed by activation of Dy^{164} and Au^{197} detectors, fixed on thin Al frames. The axial and radial flux scannings were made for a height of about 60 cm, starting from the bottom of the thimble, and across the empty sample container lowered in the thimble, respectively.

The Dy^{164} detectors were 12 mm dia. foils, punched from a 0.15 mm thick sheet made from a Dy-Al alloy containing 5% Dy by weight. The Au^{197} detectors were 12 mm dia. foils, obtained from a 0.10 mm thick sheet made from a Au-Pu alloy containing 0.1% Au by weight. The self-shielding of the Au^{197} main neutron activation resonance at 4.9 eV, corresponding to the Au surface density of the detector, was calculated to be 1.5% (33).

The detectors were irradiated either bare and contained in 1 mm thick Cd boxes. The effective cut-off energy, corresponding to the thickness and size of the Cd boxes used, was evaluated to be about 0.6 eV (25).

The induced activities of the detectors were measured by beta counting the 139 min. Dy^{165} decay, and gamma counting the 2.7 d Au^{198} decay; standard corrections for counter dead time and background, detector decay, etc., were applied to the data.

The results of these preliminary measurements are reported in Fig.15 to 17. From the analysis of the measured axial distributions it appeared that the flux maximum occurred at about 35 cm from the bottom of the thimble and that around this point the gradient of the flux was sufficiently low for the correct performance of the experiment. Consequently the length of the Al rod supporting the sample was adjusted so as to have the sample mid-plane at 35 cm from the bottom of the thimble. The analysis of the radial distribution curve permitted to verify the required flatness of the resonance flux distribution (at the 4.9 eV resonance of gold) across the Cd-clad sample container.

A.2.2. Absolute flux determination

The absolute measurement of the thermal flux, on the axis of the vertical thimble through the center of the Ispra-I core, provided the information required for the reactor safety considerations outlined in Appendix A.1., as well as for the definition of the conditions for the experiment (as reactor power, irradiation length, etc.). The measurement was performed by activating thin Au detectors and counting their induced activities by the beta-gamma coincidence technique. The procedure followed was that described by K.E.LARSSON (21), with minor modifications.

The flux detectors were 12 mm dia., 0.10 mm thick Pb-Au foils, containing 0.1% Au by weight. Such low Au concentration was chosen so as to make self-shielding, shadowing and flux perturbation effects negligible.

The foils were irradiated bare and cadmium-clad, fixed on a 70 cm long thin Al frame, lowered to the bottom of the thimble. During the exposure of the foils the reactor power was kept constant at 1.2 kW.

The induced activities of the Au-Pb foils were counted by a beta-gamma coincidence equipment, in which the radiation detectors were a 5 mm thick stilbene crystal and a 2 in. x 2 in. NaI(Tl) crystal, facing each other inside a 5 cm thick lead shield. The block diagram of the equipment is presented in Fig.18. Incidentally, the same equipment, connected to two 2 in. x 2 in. NaI(Tl) crystals, was used for the counting of the Np^{239} activity by the gamma-gamma prompt coincidence technique described in Appendix A.5.

The formulas used to infer the absolute value of the flux from the measured distintegration rates, were :

$$A = \frac{(N_\gamma - B_\gamma)(N_\beta - B_\beta - \alpha N_\gamma - \beta N_\gamma)}{(N_{\beta\gamma} - N^a_{\beta\gamma})} \qquad (10)$$

$$N^a_{\beta\gamma} = 2\tau N_\gamma \cdot N_\beta \qquad (11)$$

$$A_s = \frac{A}{1 - e^{-\lambda T}} \qquad (12)$$

$$\Phi_{th} = \frac{(A^b_s/N^b) - (A^{Cd}_s/N^{Cd})}{(g\,\sigma^\circ_{act})_{Au}} \qquad (13)$$

In the above equations the symbols have the following meaning :

$N_\gamma, N_\beta, N_{\beta\gamma}$: number of pulses recorded per unit time in the gamma, beta and coincidence channels, respectively (corrected for the equipment dead time) ;

B_γ, B_β : room background recorded per unit time in the gamma and beta channels, respectively ;

αN_γ : pulses recorded in the beta channel due to detection of electrons from internal conversion of the gamma radiation (per unit time) ;

βN_γ : gamma radiation recorded in the beta channel (per unit time) ;

$N^a_{\beta\gamma}$: accidental coincidences, recorded per unit time ;

τ : resolving time of the coincidence equipment ;

A_s : value of A corresponding to irradiation of the Au-Pb detectors untill "saturation" (infinite length irradiation) ;

λ : decay constant for Au^{198} ($1.789.10^{-4}$ min^{-1}) ;

T : irradiation length ;

Φ_{th} : thermal neutron flux ;

N : Au atom content of the Au-Pb detector ;

$(\sigma^\circ_{act})_{Au}$: activation cross section of Au for 2200 m/sec neutrons, taken equal to 98.4 b (9) ;

g_{Au} : WESTCOTT factor for Au, taken equal to 1.005 (9) ;

b, Cd : indexes for the foils irradiated bare and wrapped by cadmium, respectively.

The subtraction of the quantity αN_γ in Eq.10 was obtained experimentally, on the basis of the difference in energy between the primary betas from the decay of Au^{198} and the internal conversion electrons. To this effect, increasing thicknesses of a high purity Al absorber were interposed between the activated Au-Pb foil and the stilbene crystal, and the quantity $n = N_\gamma \cdot N_\beta / N_{\beta\gamma}$ was plotted versus the Al thickness expressed in mgr/cm^2 (Fig.19).

One obtained a reduction of about 4% in n in the interval from 50 to 100 mgr/cm^2, due to the progressive elimination of the low-energy conversion electrons, and successively a constant value of n up to the largest absorber thickness used (200 mgr/cm^2), corresponding to the detection of only the primary 0.96 meV betas of Au^{198}. Incidentally the experimentally measured conversion coefficient for Au^{198} is about 0.05. Basing on the result of this measurement, an Al absorber with a thickness of 120 mgr/cm^2, placed between the Au-Pb foil and the stilbene crystal, was used to eliminate the contribution of the conversion electrons to the pulses recorded in the beta channel ($\alpha N_\gamma = 0$).

The quantity βN_γ, namely the gamma radiation recorded in the beta channel, was determined by further increasing the thickness of the Al sheet, shielding the stilbene crystal, untill complete absorption of the betas. The accidental coincidence rate $N^a_{\beta\gamma}$ was obtained from Eq.11, using for the resolving time of the equipment (τ) the value of 0.27 μsec, determined experimentally (Fig.20).

The measured distribution of the absolute thermal flux on the axis of the central thimble of Ispra-I is shown in Fig.21. The estimated error for each experimental flux point is $\pm$5%.

A.3. TECHNIQUE FOLLOWED FOR THE PREPARATION OF THE SAMPLE FOR COUNTING

Since direct counting of the induced activities of the 4 mm thick UC detectors was not feasible, it was necessary to develop a technique for dissolving the UC disks and for obtaining from the solutions liquid samples, with precisely known amounts of U and whose activities were related to the solid detectors activities only by the corresponding weight ratios. The procedure followed for the preparation of the liquid samples for counting represented one of the most delicate aspects of the experimental technique, and is described in some detail in the following paragraphs.

The UC detectors were weighted before and after irradiation, in order to verify that no appreciable reduction in weight had resulted from chipping off the edges during the operation of loading and unloading the measuring UC rods.

The UC disks were successively calcinated at 1200°C for about 4 hours, inside a platinum crucible placed in an oven provided with an air circulation system. The outlet air was passed through an absolute filter retaining the gaseous fission products liberated in the process. The resulting U_3O_8, which had the aspect of a very fine grey powder, was weighted to check that no loss occurred during the calcination (in fact a too rapid increase in the temperature of the UC disk could result into minor explosions projecting UC fragments outside the crucible).

The U_3O_8 powder thus obtained was dissolved in a boiling acqueous solution of nitric acid. In about 10 minutes the liquid became limpid, showing that complete dissolution of the U had occurred. The cooled-off U solution was poured into a graduated flask and diluted to a total volume of 200 cc. The exact amount was checked by weighting.

After homogeneization by mixing, 3 cc of the solution were transferred, by means of a very accurate pipette, into a cylindrical teflon capsule. These capsules had been precision-machined for good-geometry gamma counting and the uniformity of their dimensions had been tested using a Palmer caliper. Again the exact amount of solution contained in each capsule was checked by weighting.

Several 3 cc counting samples were obtained from each 200 cc U solution; the U^{238} neutron capture induced activities of these samples were measured to a high statistics (following the decay of the 103 keV photopeak of Np^{239}), and only when the dispersion of the measured activities did not exceed $\pm$0.5% one of these samples was selected for counting.

The several cross-checking volume and weight determinations, carried out during the preparation of the samples for counting, coupled with the induced activities calibration, assured the precise knowledge of the amount of U contained in each sample.

A.4. EXPERIMENTAL DETERMINATION OF THE CORRECTION FOR THE SELF-ABSORPTION OF THE GAMMA RADIATION EMITTED BY THE SAMPLES

The determination of the dependence of the self-absorption of the measured 103 keV gamma rays emitted by the 3 cc liquid samples obtained from the activated UC detectors, on the U atom density, was performed using 3 cc liquid samples of varying concentrations, made from the same activated UC specimen.

Six samples were considered, with U concentrations ranging from 0.125 to 0.75 g/cm^3. The preparation of the samples was carried out as indicated in Appendix A.3. The U^{238} neutron capture induced gamma activities of the samples were counted to a high statistical accuracy, using the experimental technique and counting equipment described in the text. The irradiation times and flux levels were selected so that the above induced activities were in the range of the activities measured in the actual experiment. By this procedure one obtained the self-absorption values corresponding to the true experimental conditions.

The results of the self-absorption measurement are presented in Fig.22. The quoted errors associated with each experimental point, indicated by the flag, are the standard deviations of the mean of 4 independent runs. The measured dependence of the self-absorption on the U concentration of the sample appears to be almost linear over the entire range investigated, with a slope corresponding to a 0.2% variation per mgr/cm^3 concentration change.

Since the U concentration ranges of the actual 3 cc samples were 0.48 to 0.51 $mgr/_3cm^3$ for the 25.2 mm UC detectors, and 0.73 to 0.76 $mgr/_3cm^3$ for the 30.9 mm UC detectors, the correction factors applied to the measured activation ratios were always close to unity, and the uncertainty introduced in the data by this correction was negligible.

A.5. EXPERIMENTAL DETERMINATION OF THE CORRECTION FOR THE FISSION PRODUCT DECAY ACTIVITY OF THE SAMPLES

The correction for the fission product activity contamination (from U^{235} epicadmium fission and U^{238} fast fission) in the energy range scanned, was determined by monitoring the activity of the 3 cc sample above 410 keV (the Au^{198} main gamma ray, chosen for convenience), i.e. where the activity a few hours after the end of the irradiation is only from the fission product decay, and reducing it to the corresponding fission product activity present in the energy interval of interest by means of a normalization factor obtained experimentally.

To this effect, a U-Al foil, containing U enriched to 90% U^{235}, was irradiated in a neutron flux, and the induced fission product activity of a 3 cc liquid sample obtained from the foil was measured in the 50 keV interval about 103 keV (f_{103}) and integrally above 410 keV (f_{410}), respectively.

If F_{410} is the fission product activity of the U^{238} resonance detector above 410 keV, then the fission product term to be subtracted from the measured total activity of such detector in the 50 keV interval around 103 keV is $F_{103} = (f_{103}/f_{410}) \cdot F_{410}$. The principle of the correction is clarified in Fig.23, showing the measured gamma rays spectra from the decay of the U and U^{235} samples, as well as the energy ranges considered.

The value of the normalization factor (f_{103}/f_{410}) is in general a function of neutron spectrum, irradiation length, time elapsed after the irradiation, counting technique, counting equipment settings, etc.; in the present case the effect of these parameters was cancelled out by irradiating a U^{235} foil together with each rod sample, and

counting its activity together with the activity induced in the corresponding U^{238} resonance detector.

In the actual experimental set-up, the output pulses from each of the two photomultipliers were fed to two amplifier-discriminators, set to the best operating conditions in the regions around 100 keV and above 400 keV, respectively, in turn feeding two six-decades fast scalers.

Although the principle of the fission product correction method using U^{235} - Al foils was not rigorously correct (mainly because the fission product distributions from U^{235} and U^{238} fissions are not exactly the same, and the self-absorption of the emitted gamma radiation was not the same for the U^{235} - Al and U liquid samples), the application of the technique to the present case was justified by the low values of the fission product activity contaminations (1-2% of the measured total activities), which were also approximately the same for the UC reference rods and the UC cluster rods activations.

In some instances, an independent technique for selectively determining the U^{238} neutron capture induced activities of the samples was used, consisting in measuring the 2.3^{d} half-life decay coincidence (34,35) between the 106 keV gamma ray and the 103 keV internal conversion X-ray from the deexcitation of Pu^{239}. Small corrections were applied to the measured coincidence rates for the U^{238} natural activity, for the minor residual fission products activities contamination - as determined by a procedure analogous to that outlined above - and for accidental coincidences.

Being rather cumbersome, this technique was utilized only as a test for the method described above, essentially to check the validity of the fission product activity correction procedure. Basing on the very good agreement between the ratios of the U^{238} resonance capture activities induced in the UC clusters and in the UC reference rod, obtained by the two techniques used, it was concluded that the uncertainty of the fission correction could not introduce more than a 0.2% error into the results.

A.6. MEASUREMENT OF THE EFFECTIVE RESONANCE INTEGRAL OF A U ROD BY REFERENCE TO A AU STANDARD

At the start of the project, a measurement of the effective resonance integral of a 12 mm dia. U rod - by reference to the infinite dilution resonance integral of gold - was performed in the central thimble facility of the Ispra-I reactor.

Purpose of this preliminary experiment was to test the equipment and the technique set up for the programmed U^{238} resonance capture studies, as well as to provide indirect evidence for the assumed close-to-1/E dependence of the slowing-down neutron flux in the center of the Ispra-I core.

A.6.1. Details of the measurement

Basically, the technique used consisted in comparing the resonance neutron capture activities of a U rod sample and of an extremely thin Au foil sample, irradiated under equal conditions in a neutron slowing-down flux. The data were normalized to the ratio of the activities of the same detectors, when irradiated in a Maxwellian neutron spectrum.

The U rod sample consisted of an Al-clad, 12 mm dia., 30 cm long split-rod, containing a 0.1 mm thick U foil, depleted to 350 ppm U^{235}. The Au sample was a 12 mm dia., 0.1 mm thick Au-Pb foil, containing 0.1% Au by weight.

The U and Au samples were irradiated in turn, inside the Cd-clad container designed for the 19-rod U cluster measurements placed at the core mid-plane in the central thimble of the Ispra-I reactor. The irradiations were carried out at a nominal reactor power of 1.2 kW, for 20 minutes.

The resonance flux was monitored by 12 mm dia., 0.2 mm thick pure Au detectors, contained inside a 0.8 mm thick Cd box, and fixed to the Al supporting rod of the sample holder, at a distance of 20 cm from the upper end of the rod sample. This distance was determined so as to insure a negligible shielding of the 4.9 eV resonance of the gold monitor by the lowest U^{238} resonance of the U rod sample. For the justification of the use of this "unperturbed flux" monitoring technique one is referred to Section 2 in the text.

The irradiations in the Maxwellian flux were carried out using the thermal column of the Ispra-I reactor, operated at 50 kW power. The samples consisted in sandwiches of one Au-Pb foil between two depleted U foils, including 0.01 mm thick pure Al catchers, to prevent contamination of the Au-Pu foils by the fission products emitted from the U foils surface.

After irradiation, the U^{238} neutron capture induced activities of the U foils were measured by the β-γ coincidence technique outlined in Appendix A.5. The Au induced gamma activities were measured by following the 410 keV, 2.7 d half-life decay of Au^{198}.

According to the experimental technique outlined above, the effective resonance integral of the U rod sample, RI_U, is inferred from the measured quantities by means of the expression (6) :

$$k \cdot RI_U - \sigma_U^{1/v} = \left\{ \frac{A_U^{res}}{A_{Au}^{res}} \cdot \frac{A_{Au}^{th}}{A_U^{th}} \right\} \left\{ \left(RI_{Au} - \Delta RI_{Au} - \sigma_{Au}^{1/v} \right) \frac{(g\sigma_0)_U}{(g\sigma_0)_{Au}} \right\} \quad (14)$$

$$= \{A\} \cdot \{B\}$$

where A is the measured dimensionless parameter, and the quantity B groups cross sections and other nuclear data obtained from the values published in the literature.

In Eq.14 the symbols have the following meaning :

A_U^{res}, A_{Au}^{res} : the saturated activities of the U and Au-Pb foils irradiated in the slowing-down spectrum, normalized to the corresponding saturated activities of the Au monitors ;

A_U^{th}, A_{Au}^{th} : the saturated activities of the U and Au-Pb foils irradiated in the Maxwellian flux ;

RI_{Au} : infinite dilution resonance integral of Au^{197} ;

ΔRI_{Au} : correction to the measured resonance capture in the Au-Pb foil, due to the self-shielding of the 4.9 eV resonance of Au^{197} (33) ;

$\sigma_U^{1/v}$, $\sigma_{Au}^{1/v}$: contribution of the 1/v varying part of the cross section to the neutron capture at energies above the Cd cut-off in the U^{238} and Au^{197} foils, respectively ;

$(g\sigma_o)_U$, $(g\sigma_o)_{Au}$: the 2200 m/sec cross sections, corrected for deviations from 1/v in the thermal region, of U and Au, respectively (following the WESTCOTT formalism (36)) ;

k : correction factor taking into account the departure of the actual slowing-down flux, from the ideal 1/E dependance required by the definition of effective resonance integral.

The values assumed for the nuclear constants in Eq.14 are listed in the Table below :

Symbol	Absolute Value	Reference
RI_{Au}	1510 ± 30 barns	9
ΔRI_{Au}	20 ± 5 "	32
$\sigma_{Au}^{1/v}$	40 ± 5 "	9
g_{Au}	1005 (T=20°C)	9
$(\sigma_o)_{Au}$	98.4 ± 0.5 "	9
$\sigma_U^{1/v}$	1.2 ± "	6
g_U	1002 (T=20°C)	6
$(\sigma_o)_U$	2.71 ± "	6

A.6.2. Test of the slowing-down neutron spectrum in the Ispra-I reactor

Basing on the experimental data and Eq.14, an indirect test of the magnitude of the departure from the 1/E behaviour of the slowing-down neutron flux in the central thimble facility of the Ispra-I reactor (or more precisely of the effect of such departure on the measured effective resonance integrals) was performed, as explained below.

Eq.14 was solved for k, yielding :

$$k = \frac{\{A\}\cdot\{B\} - \sigma_U^{kv}}{RI_U} \qquad (15)$$

The correction factor k depends on the true slowing-down spectrum in the experimental facility, as well as on the resonance energy ranges of the sample, the standard and the monitor (in the actual case : respectively U, Au and Au). Provided one compares two experiments performed using the same technique (i.e. also with the same standard, monitor, etc.), the difference in the corresponding values of k is a function only of the difference in the experimental slowing-down neutron spectra.

A very accurate measurement of the effective resonance integral of U and UO_2 rods was performed by E.HELLSTRAND, by the same method used for the test outlined above, in the D_2O moderated R-1 reactor (6). The neutron slowing down flux in the experimental facility of such reactor was measured using a fast chopper (12,13), showing a slight deviation from

the 1/E dependence, which corresponded to a correction factor $k_{(R-1)} = 1.04 \pm 0.02$.

The results of the measurement for U were summarized in the expression :

$$RI_U = 2.95 + 25.8\sqrt{S/M} \qquad (16)$$

valid in the range $0.07 < S/M\ (cm^2/g) < 0.53$. According to Eq.16 the value of the effective resonance integral of a 12 mm dia. ($S/M = 0.18\ cm^2/g$) U rod is 13.79 barns.

By introducing such calculated value of RI_U into Eq.15, giving to the parameter B the value obtained using the cross section data listed in the Table above and to the parameter A the value obtained from the present measurement, one got :

$$k_{(Ispra-I)} = 1.07 \pm 0.04$$

The relatively close agreement between the values of the spectral corrections factors for R-1 and Ispra-I (1.04 ± 0.02 vs. 1.07 ± 0.04) supported the starting assumption of the similarity in the slowing-down neutron spectra in the two D_2O moderated reactors. The conclusion drawn from this test, was that the slowing-down neutron flux at the experimental position in Ispra-I was sufficiently close to the 1/E dependence, for the technique selected for the experiment described in the text to be valid (see Section 2 of the report).

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coolant	experimental values: boundary resonance flux monitoring R_1	RI_1 (barns)	average resonance flux monitoring R_2	RI_2 (barns)	calculated values R
diphyl (with graphite matrix)	0.947 ±0.012	10.7 ±0.5	0.977 ±0.020	11.1 ±0.6	0.990
air (with graphite matrix)	0.920 ±0.013	10.4 ±0.5	0.938 ±0.015	10.6 ±0.5	0.979
diphyl (without graphite matrix)	-	-	0.986 ±0.010	11.25 ±0.6	0.968

Table 1 : Experimental results for R and RI and calculated values for R.
Four-rod UC clusters

coolant	experimental values: boundary resonance flux monitoring R_1	RI_1 (barns)	average resonance flux monitoring R_2	RI_2 (barns)	calculated values R
air	0.794 ±0.008	9.6 ±0.5	0.838 ±0.008	10.2 ±0.5	0.801
D_2O	0.790 ±0.011	9.5 ±0.5	-	-	-
diphyl (with Mg fillers)	0.822 ±0.008	10.0 ±0.5	0.881 ±0.009	10.8 ±0.5	-
diphyl (without Mg fillers)	0.874 ±0.019	10.7 ±0.5	0.906 ±0.021	11.1 ±0.6	0.894
H_2O	0.820 ±0.017	10.0 ±0.5	0.893 ±0.008	10.95 ±0.5	0.915

Table 2 : Experimental results for R and RI and calculated values for R.
Seven-rod UC clusters

coolant	average resonance flux monitoring		boundary resonance flux monitoring	
	center rod	outer rod	center rod	outer rod
air	8.6 ±0.4	10.5 ±0.5	7.8 ±0.4	9.9 ±0.5
D_2O	-	-	7.9 ±0.4	9.8 ±0.5
diphyl (with Mg fillers)	9.6 ±0.5	11.0 ±0.6	8.1 ±0.4	10.4 ±0.5
diphyl (without Mg fillers)	9.5 ±0.5	11.4 ±0.6	8.3 ±0.4	10.5 ±0.5
H_2O	10.7 ±0.5	12.1 ±0.6	7.9 ±0.4	10.3 ±0.5

Table 3 : Measured values for the effective resonance integral of each representative rod of the 7 rod UC cluster (in barns)

coolant	boundary resonance flux monitoring		average resonance flux monitoring	
	R_1	RI_1	R_2	RI_2
air	0.794 ±0.008	9.6 ±0.5	0.838 ±0.006	10.2 ±0.5
D_2O	0.839 ±0.008	10.2 ±0.5	-	-
diphyl+ filler	0.903 ±0.005	11.1 ±0.6	0.924 ±0.031	11.4 ±0.6
diphyl- filler	0.973 ±0.008	12.0 ±0.6	-	-
H_2O	1.06 ±0.01	13.2 ±0.7	1.17 ±0.015	14.7 ±0.7

Table 4 : Results of the measurements performed with the cluster rods containing the UC detectors bare.
Seven-rod UC clusters

cluster	coolant	R_1	RI_1
4 UC	diphyl (C matrix)	0.826 ±0.008	9.2 ±0.5
4 UC	air (C matrix)	0.823 ±0.012	9.2 ±0.5
4 UC	diphyl (without C matrix)	0.855 ±0.010	9.6 ±0.5
7 UC	air	0.761 ±0.005	9.1 ±0.5
7 UC	D_2O	0.732 ±0.014	8.8 ±0.4
7 UC	diphyl (with Mg fillers)	0.722 ±0.008	8.6 ±0.4
7 UC	diphyl (without Mg fillers)	0.690 ±0.007	8.2 ±0.4
7 UC	H_2O	0.612 ±0.007	7.1 ±0.4

Table 5 : Experiments results for R_1 and RI_1, obtained using the "unperturbed flux" monitoring technique.

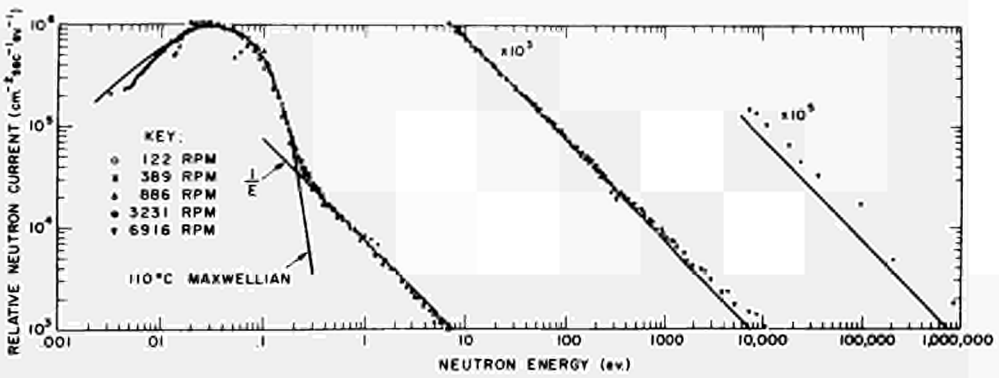

Fig. 1. Leakage Neutron Energy Spectrum, Third Series of Experiments. Two Fuel Elements in Front of 12-in. Port.

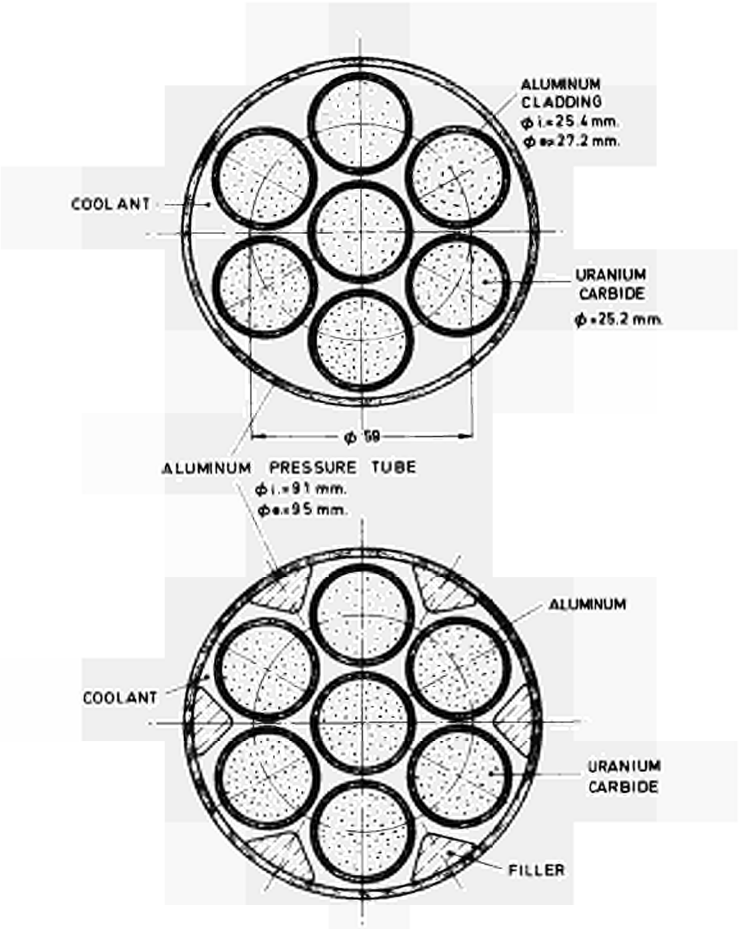

Fig. 2 GEOMETRY OF THE 7-ROD UC CLUSTER

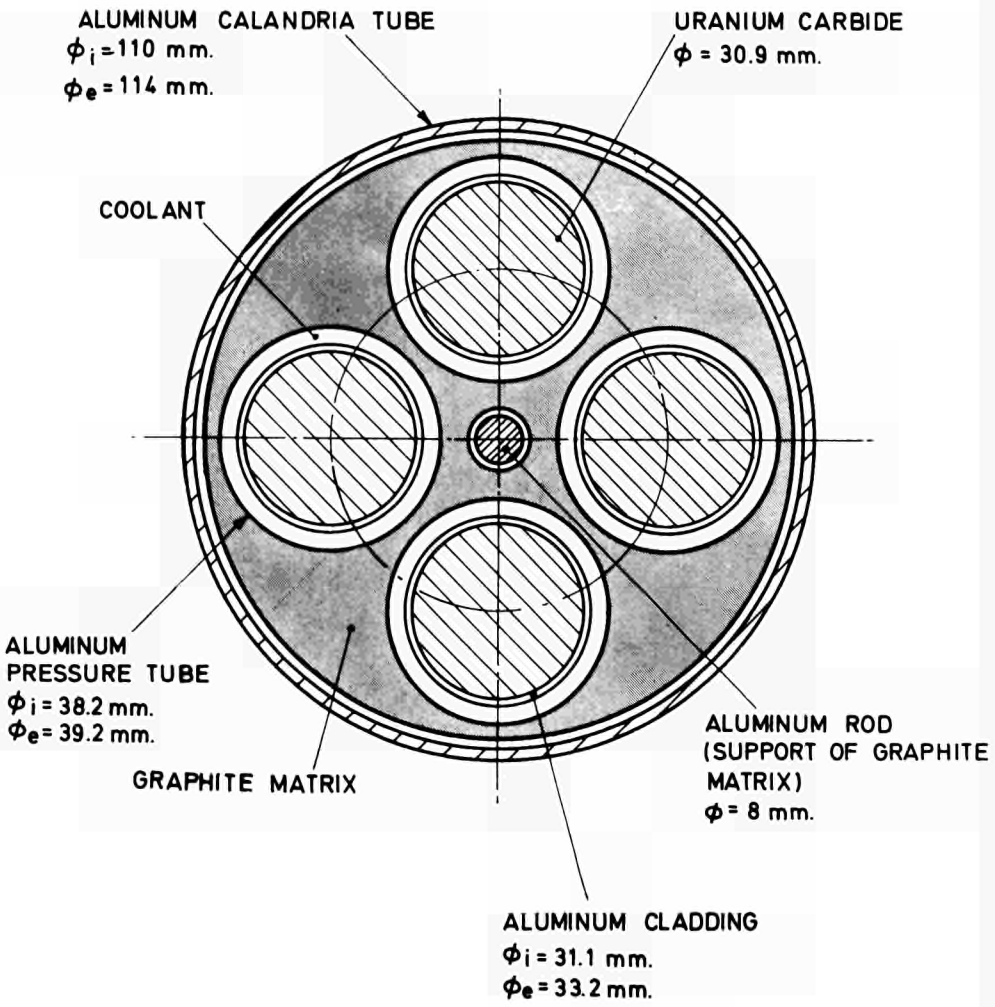

Fig. 3 GEOMETRY OF THE 4-ROD UC CLUSTER

Fig. 5 : Experimental Assembly. Assembly ready for irradiation, with details of support system.

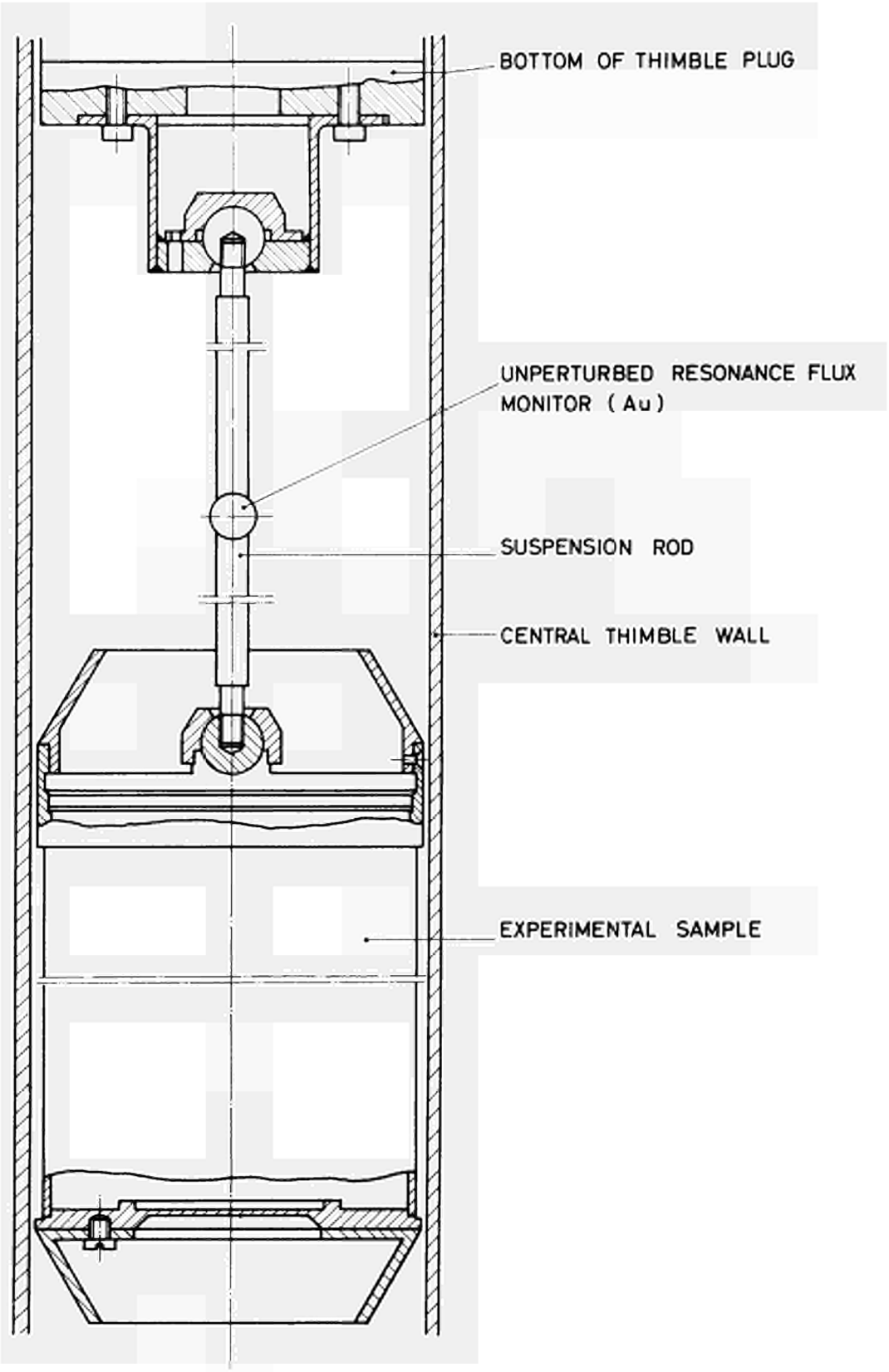

Fig. 4 VIEW OF THE EXPERIMENTAL ASSEMBLY IN THE CENTRAL THIMBLE OF ISPRA - I

Fig. 6 : Experimental Assembly. Details of the 7-rod cluster sample (from top right : Cd-clad container with UC rods, outer container, UC rod clad by Al with Cd sheet around the central section, lid of inner container with safety pressure valve and expansion ring, Cld lid, lid of external container with support rod provided with spherical joint)

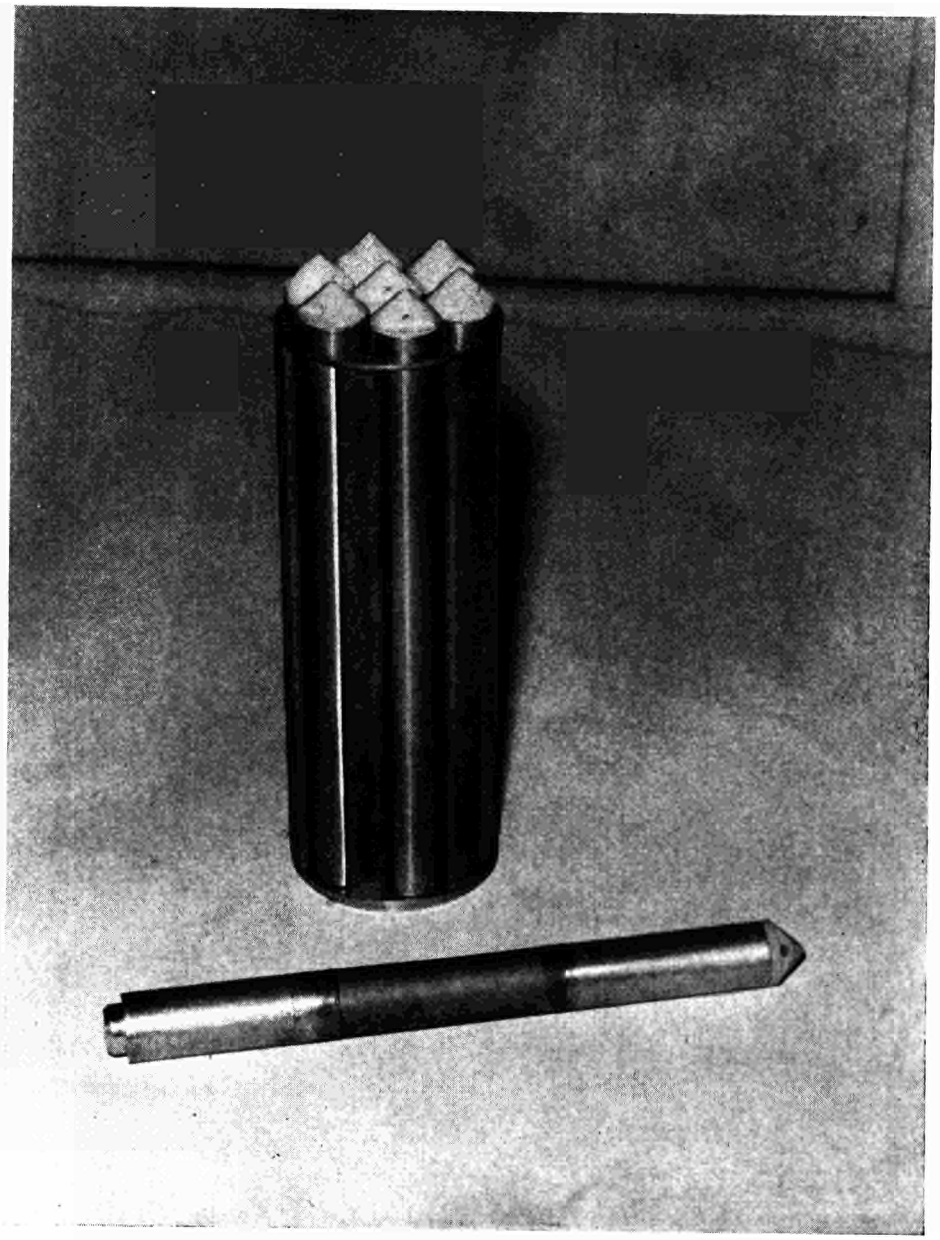

Fig. 7 : Experimental Assembly. Assembly of 7-rod cluster with Mg fillers.

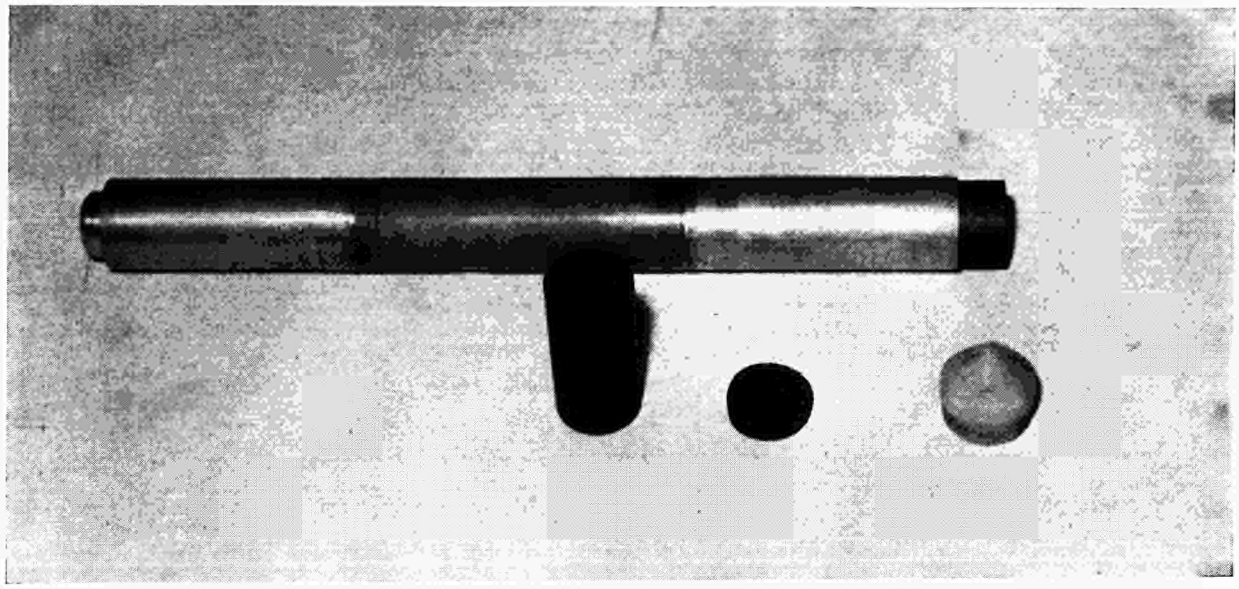

Fig. 8 : Experimental Assembly. Details of 25.2 mm dia. UC rod (from left : Al cladding with Cd sheet around central section, UC pellet, UC detector, teflon lid to cladding).

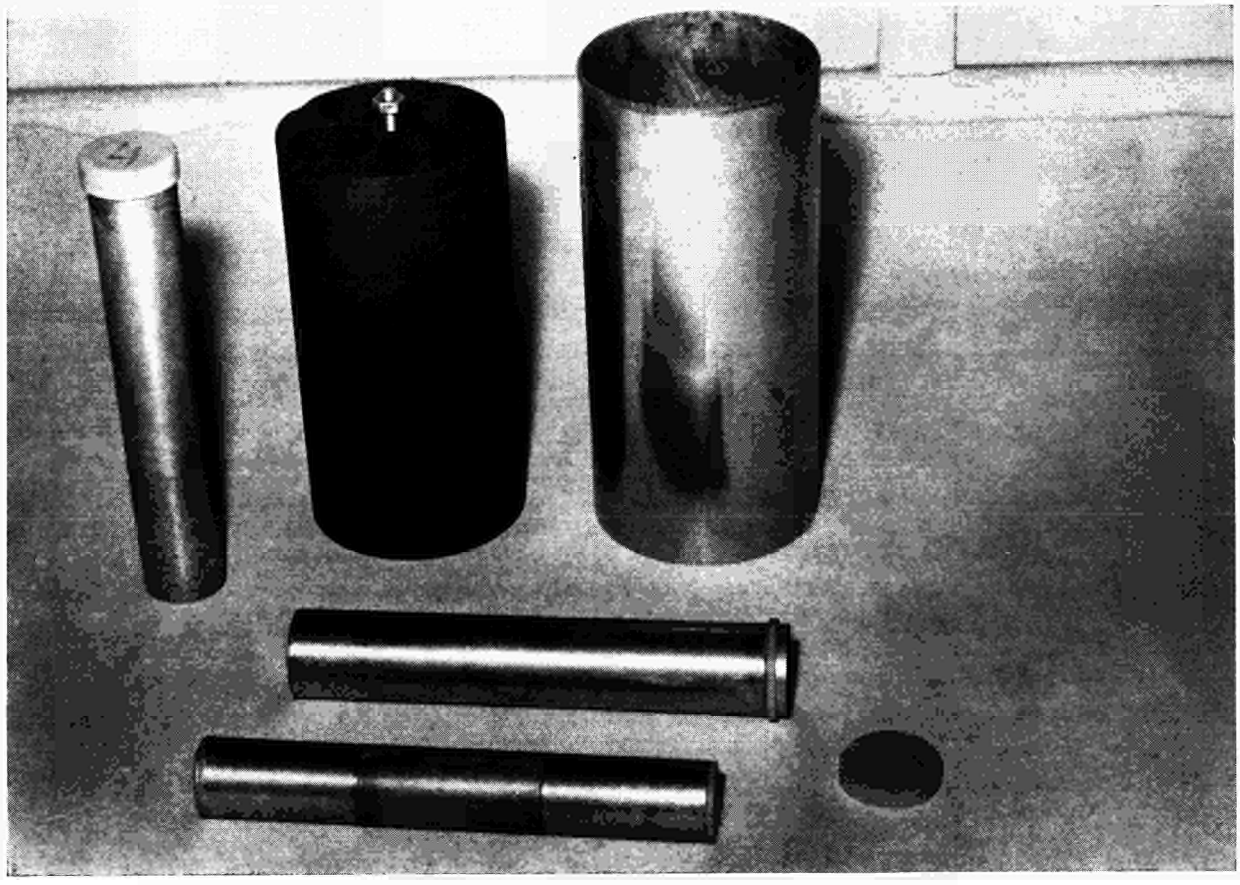

Fig. 9 : Experimental Assembly. Details of 4-rod cluster sample (from top left : UC rod-pressure tube assembly, graphite matrix, Cd-clad container, Al pressure tube, UC rod clad by Al).

Fig. 10 : Experimental Assembly. Assembly of 4-rod cluster without graphite matrix.

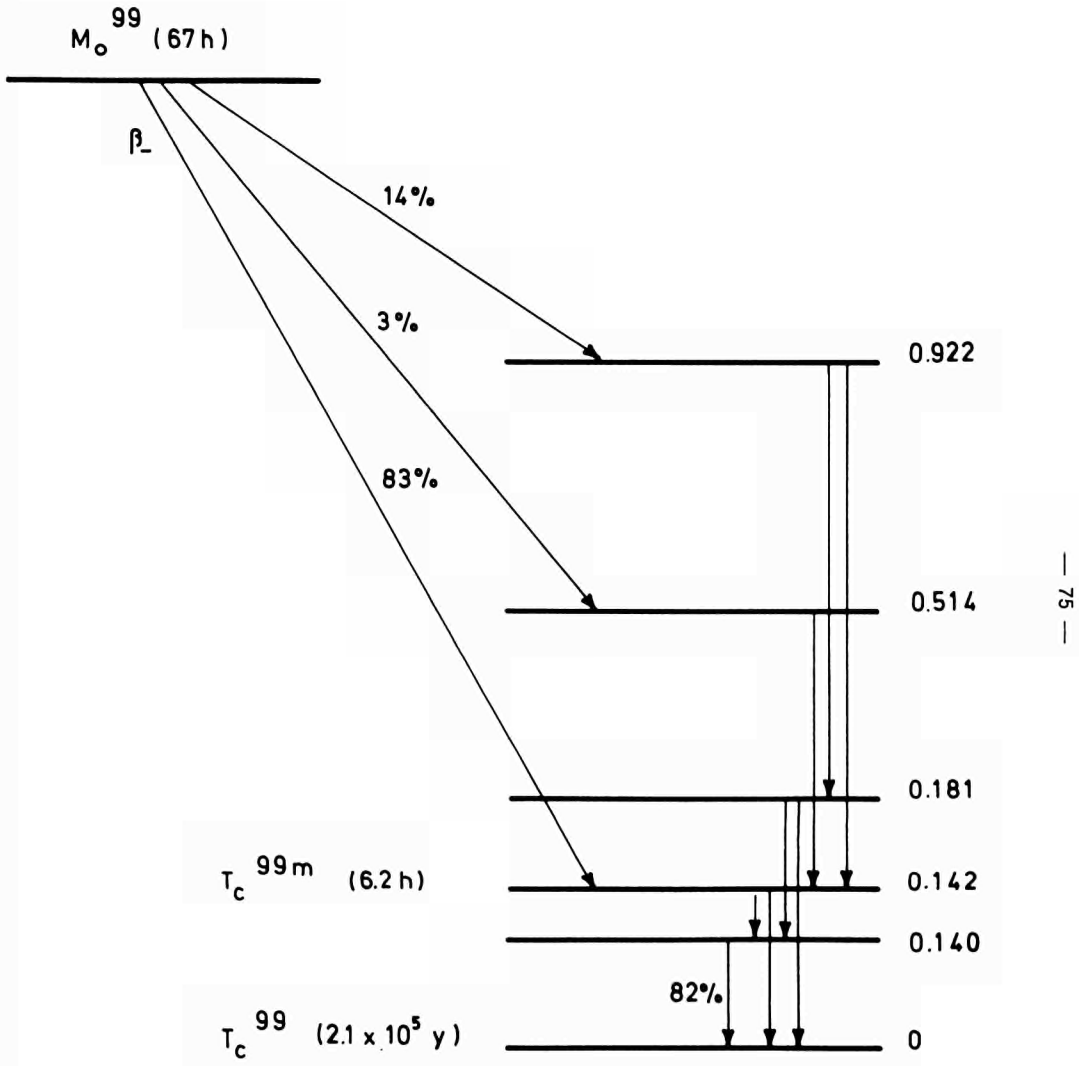

Fig. 11 DECAY SCHEME OF M_o^{99}

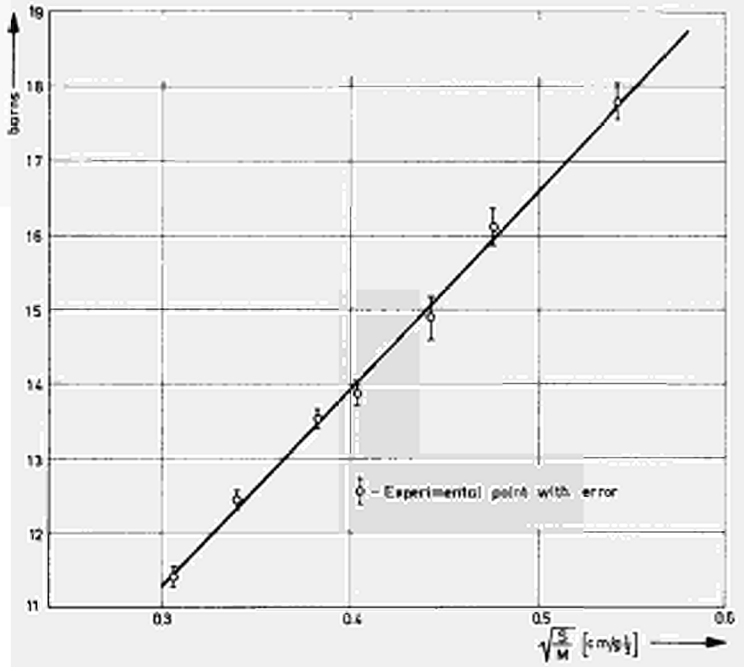

Fig.12 EFFECTIVE RESONANCE INTEGRAL OF URANIUM CARBIDE RODS

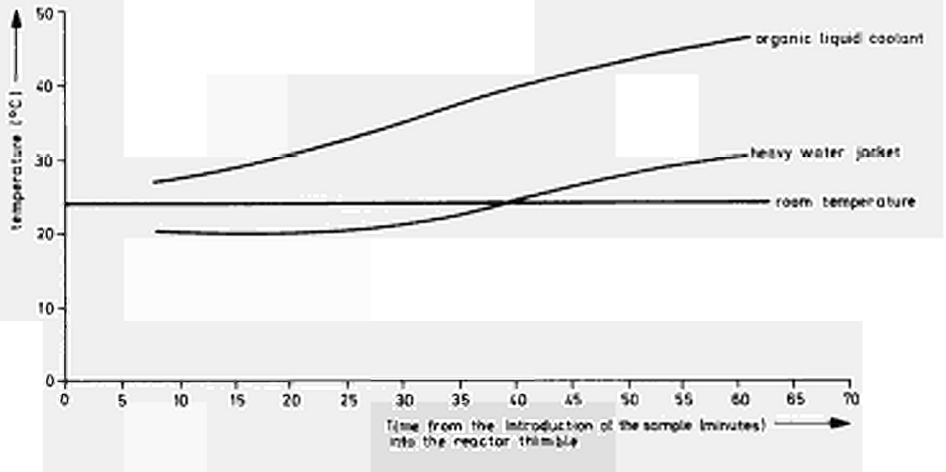

Fig.13 VARIATION OF THE TEMPERATURE INSIDE THE EXPERIMENTAL FUEL ASSEMBLY IN THE ISPRA-1 THIMBLE (REACTOR SHUT-DOWN)

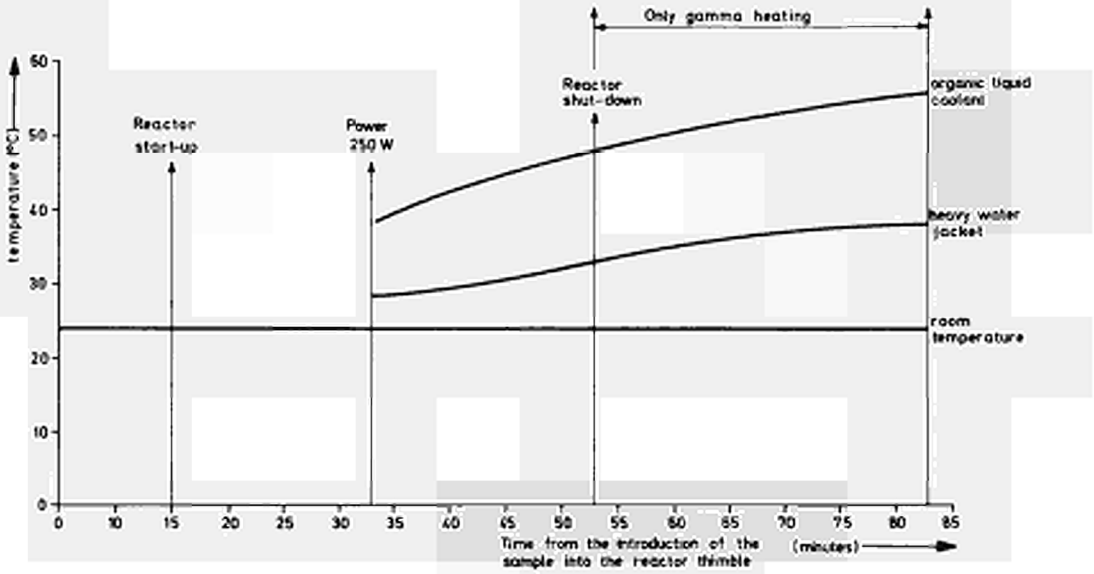

Fig. 14 VARIATION OF THE TEMPERATURE INSIDE THE EXPERIMENTAL FUEL ASSEMBLY IN THE ISPRA-1 THIMBLE (REACTOR IN OPERATION)

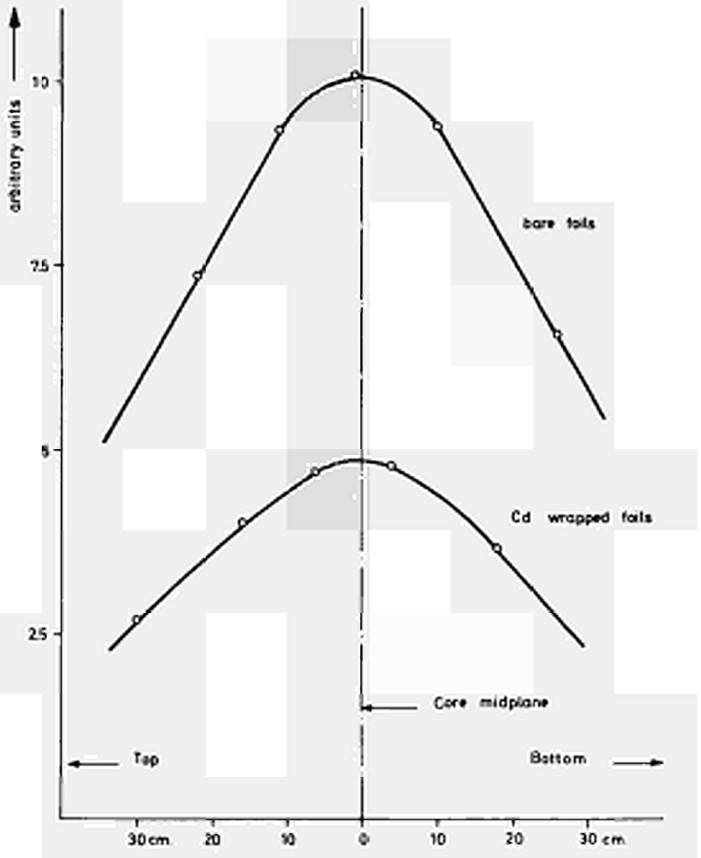

Fig. 15 AXIAL DISTRIBUTION OF THE ACTIVITY OF Au-Pb DETECTORS IRRADIATED IN THE CENTRAL THIMBLE OF ISPRA-1

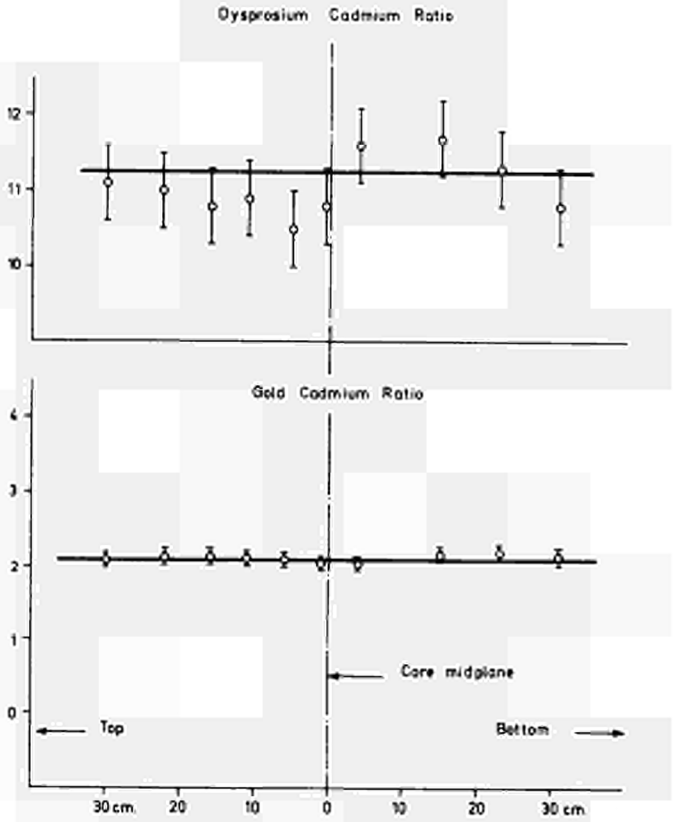

Fig.16 AXIAL VARIATION OF CADMIUM RATIOS IN THE CENTRAL THIMBLE OF ISPRA-1

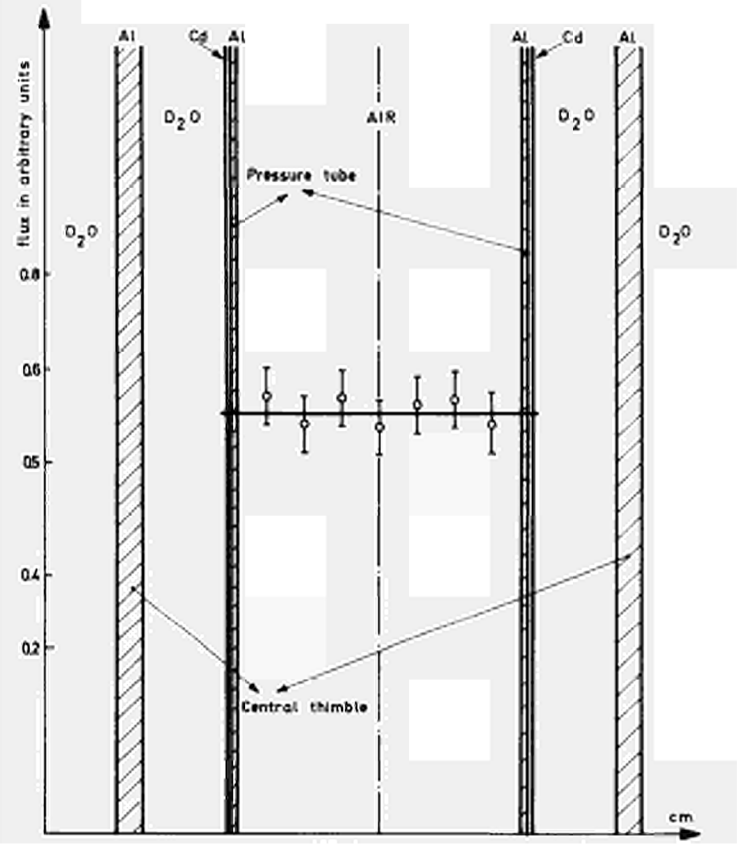

Fig.17 RADIAL DISTRIBUTION OF THE RESONANCE FLUX INSIDE THE Cd-CLAD SAMPLE CONTAINER IN THE CENTRAL THIMBLE OF ISPRA-1 (ACTIVITY OF Au-Pb DETECTORS)

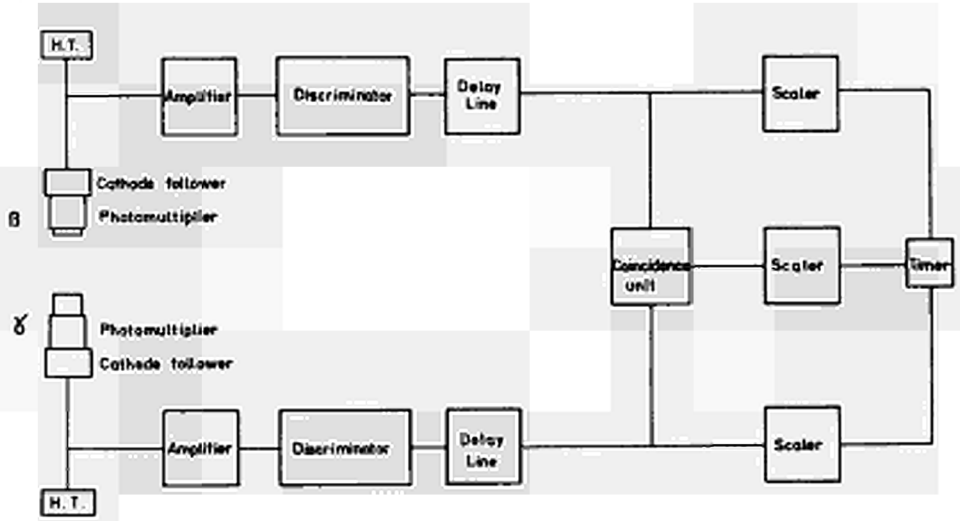

Fig. 18 BLOCK DIAGRAM OF THE β - γ COINCIDENCE SET

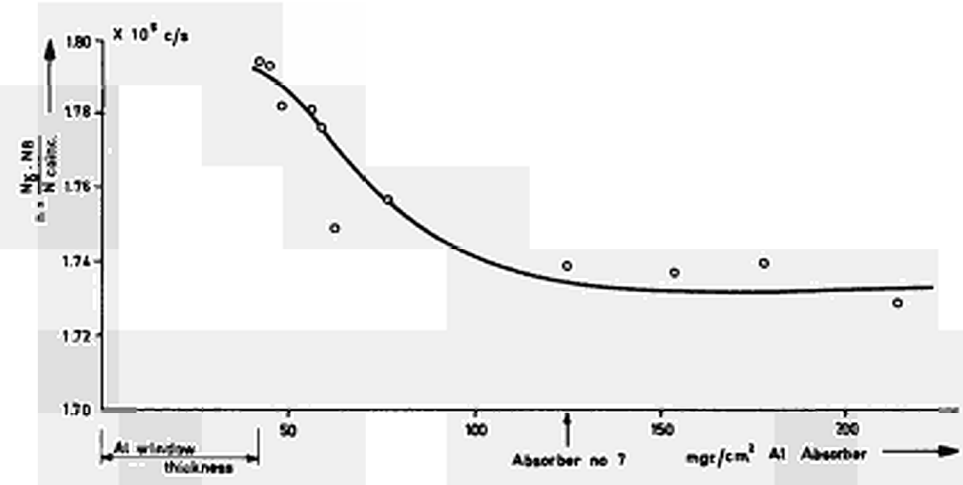

Fig. 19 $n = \frac{N\gamma - N\beta}{N\text{coinc.}}$ AS A FUNCTION OF THE ALUMINUM ABSORBER THICKNESS

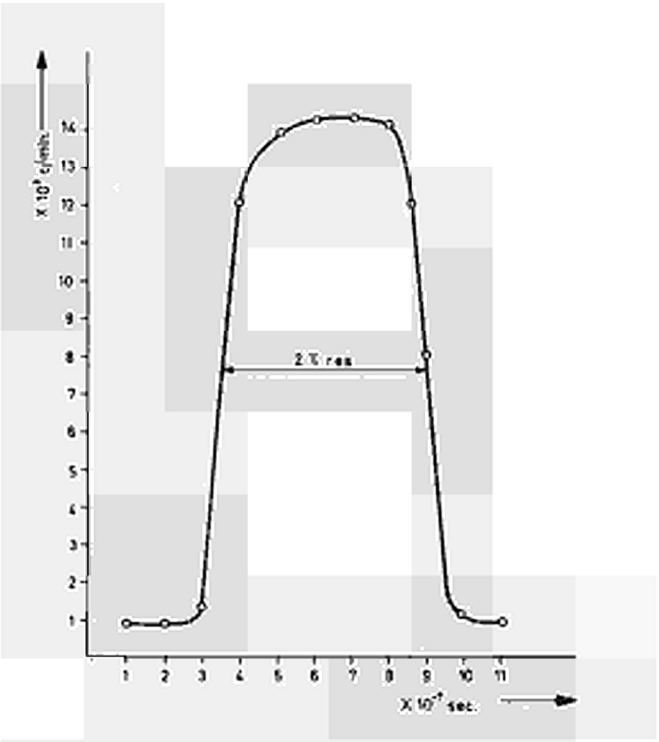

Fig. 20 DETERMINATION OF RESOLVING TIME OF COINCIDENCE EQUIPEMENT

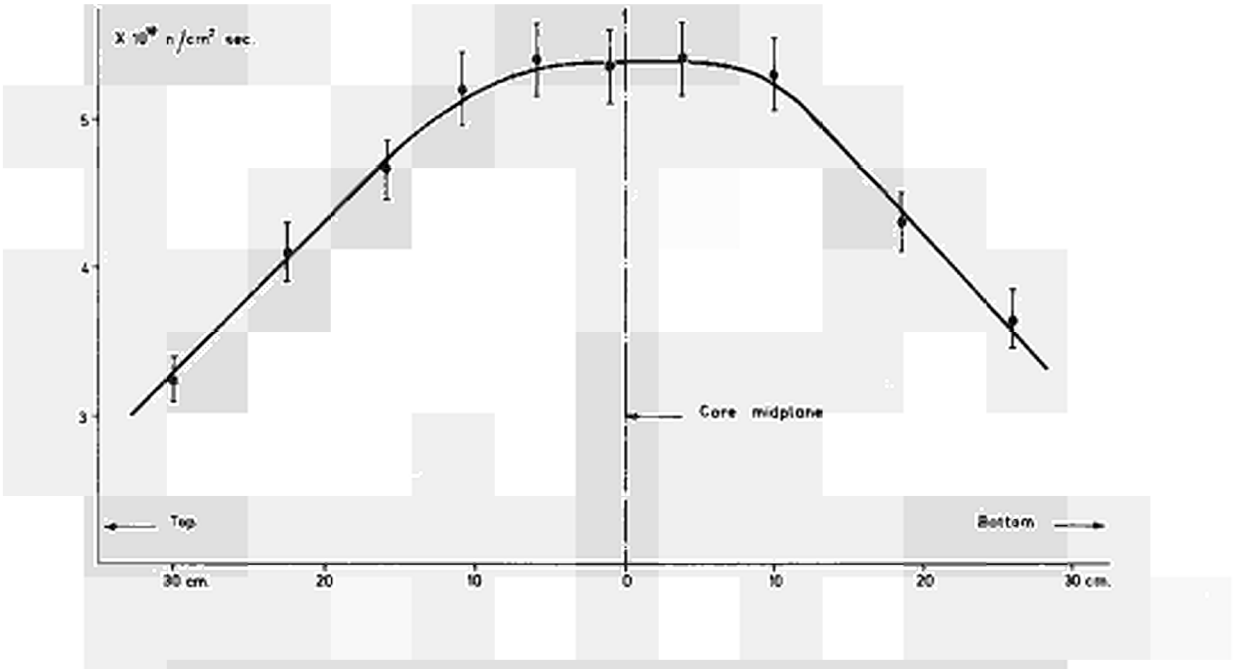

Fig. 21 ABSOLUTE THERMAL FLUX DISTRIBUTION ON THE AXIS OF THE CENTRAL THIMBLE OF ISPRA-1 AT POWER OF 1.2 KW.

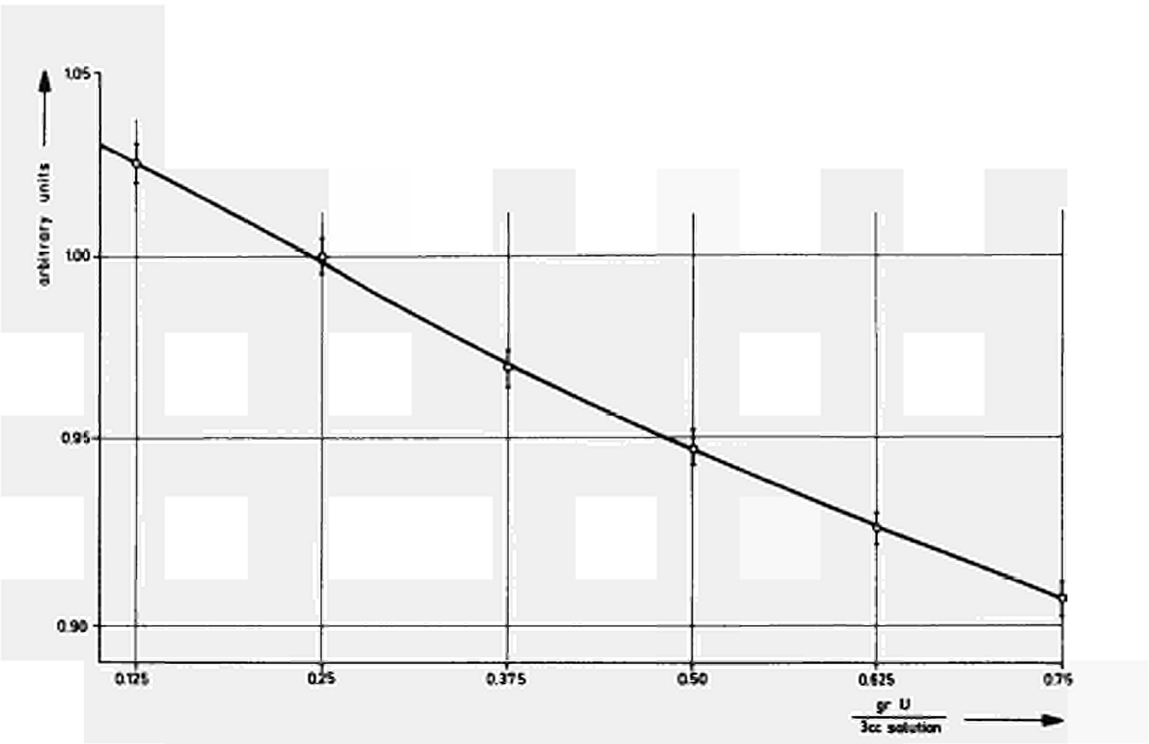

Fig. 22 SELF ABSORPTION CORRECTION AS FUNCTION OF THE URANIUM CONTENT IN 3 cc. OF SOLUTION

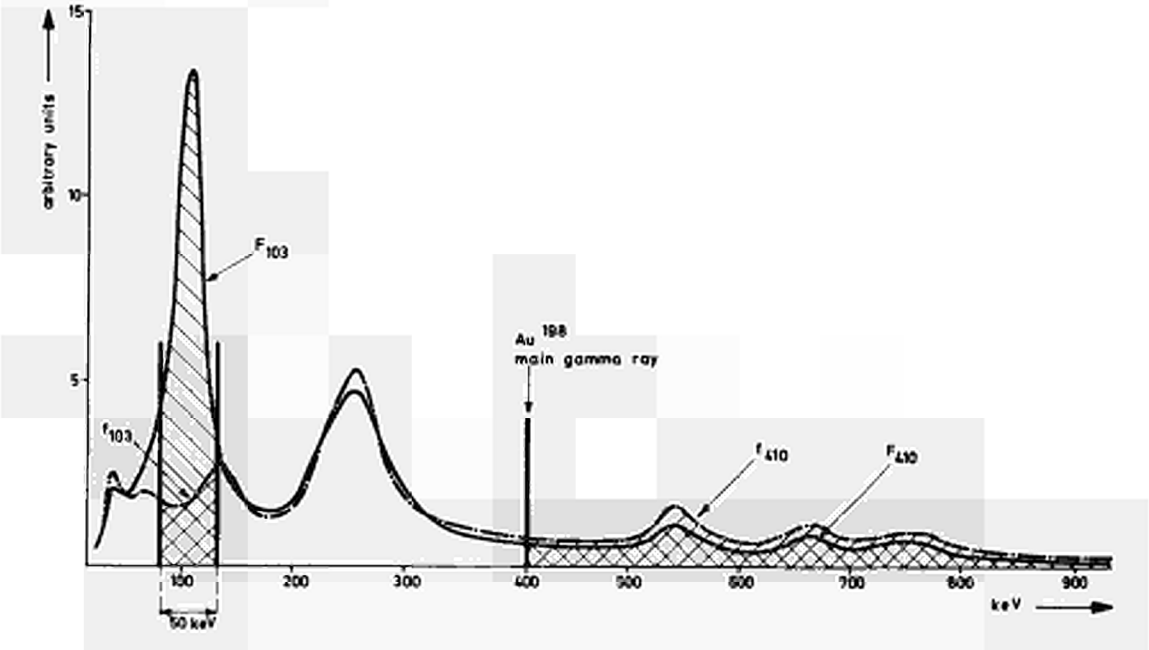

Fig. 23 MEASURED GAMMA RAYS SPECTRA FROM THE DECAY OF THE ACTIVATED NATURAL URANIUM AND U^{235} SAMPLES

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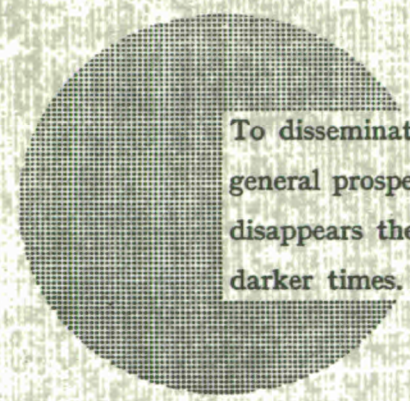

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Alfred Nobel

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