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BURNUP DETERMINATION OF NUCLEAR FUELS

Annual Report 1965

Edited by
M. BRESESTI

1967

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ORGEL Program

**Joint Nuclear Research Center
Ispra Establishment - Italy**

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Annual Report 1965

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M. BRESESTI

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Annual Report

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SUMMARY

This report describes the present state of the work of developing reliable methods for burnup determinations in irradiated uranium carbide (UC) fuel specimens to meet the requests of the Orgel Project. Several methods have been investigated, in order to be able to perform burnup measurements in fuels having different compositions and different irradiation histories. The four main methods are:

- 1) Measurement of U235 depletion.
- 2) Measurement of fission products.
- 3) Measurement of plutonium.
- 4) Measurement of neutron flux.

In order to utilize these methods it is necessary to develop techniques for isotopic composition and activity measurements and to develop radiochemical procedures. Moreover the processing of the experimental data normally requires a knowledge of the spatially dependent neutron spectra existing during the irradiation so that reactor physics calculations must be performed.

Calculations of neutron spectra and related parameters T , r , β , R (Section 1), for the UC specimens to be irradiated in the DIRCE loop of the Ispra I reactor, have been made using nuclear codes. The ratio $\bar{\phi}/\phi_s$, between the average flux in the fuel and the surface flux, has also been calculated, this value being necessary in the burnup determination starting from neutron flux measurements on the surface of the irradiated fuel.

Investigations have been performed (Section 2) concerning the preparation of samples for the different determinations starting from big, highly radioactive UC specimens as received after mechanical dismantling of the test elements. The main problem is the preparation of representative small samples starting from a complete cross-section of a UC rod.

For the measurement of U235 depletion, experiments utilizing optical spectrometry (Section 3) and activation analysis (Section 4) have been performed. For optical spectrometry the utilization of a hollow cathode lamp has allowed one to obtain much higher resolution than with a direct current arc discharge. The utilization of activation techniques for the measurement of U235 content in depleted uranium has shown that these techniques allow one to determine the U235 content with a precision of 1-2%.

Experimental results are also reported for the quantitative determination of uranium by colorimetric and coulometric methods (Section 5) and for the purification of uranium from fission products (Section 6). The quantitative determination of uranium is necessary because the ratio between fission product or plutonium activity to uranium weight is the value required for the burnup calculation.

The determination of fission products by γ -spectrometry, without any chemical separation, has been attempted utilizing NaI(Tl) and Germanium Li-drifted detectors (Section 7). Gamma spectrometry using Germanium Li-drifted detectors has allowed one to measure the activities of the fission products Cs137, Ce144 and Ru106 in irradiated samples of UC. Due to the strong difference in the fission yields of Ru106 for U235 and Pu239 fission it was possible to determine, by comparison of the Cs137, Ce144 and Ru106 activities, the separate contributions of the U235 and Pu239 to the total number of fissions.

In Section 8 the radiochemical procedure required for the purification of the stable fission product Nd148 is presented.

The Pu239 + Pu240 activity (Section 9) has been measured by α spectrometry using a silicon surface barrier detector. An isotope dilution technique was utilized in this measurement.

For the neutron flux measurements (Section 10) very dilute alloys of cobalt (1% and 0.1%) and silver (0.1%) in aluminium have been developed. The utilization of the cobalt-silver pair allows one to determine the thermal flux and the $r \sqrt{\frac{T}{T_0}}$ factor. In this connection a new determination of the s_0 factor for Ag109 has been made yielding 18.23 ± 0.36 .

In conclusion, it can be stated that 4 different independent methods for burnup determination have been set up, which can be utilized according to the irradiation history of the sample and the information wanted.

INTRODUCTION

After preliminary discussions in October 1964, it was decided that the Chemistry Department set up methods for the determination of burnup of UC specimens to be irradiated in the Ispra I reactor and in the HFR at Petten by the Metallurgy Laboratory.

In the DIRCE loop of the Ispra I reactor, irradiations of a maximum period of a few weeks in a flux of about 5×10^{13} n/cm²-sec are planned. In the HFR at Petten, irradiations of 5000 - 10000 MWd/T are planned.

During 1965, the Physical Chemistry Laboratory has required burnup analysis of UC samples irradiated in the central position of the Ispra I reactor.

An analysis of the methods reported in the literature for the burnup determination has been made, keeping in mind the characteristics of the irradiations to be studied and the equipment and experience available at Ispra. The most widely utilized methods for the determination of burnup of uranium fuels are the following:

1) Measurement of U235 depletion.

The measurement of U235 depletion allows one to determine directly the number of U235 fissions. However, this leaves the number of Pu239 fissions to be calculated. The Pu239 fission is important mainly in natural uranium irradiations. The measurement of the U235 depletion is normally made by mass spectrometry.

2) Fission product determination.

The fission products used for the measurement are normally long half life isotopes or stable isotopes. In using radioactive nuclides, sources of error are the inaccuracy in the knowledge of the nuclear data such as fission yield, decay constant, decay scheme, capture cross section, etc, and in the knowledge of the history of the irradiation. In the determination of radioactive nuclides, γ -spectrometry techniques are normally utilized.

The measurement of a stable fission product eliminates the inaccuracy due to the knowledge of the decay constant, decay scheme and history of the irradiation. The stable isotope that appears most interesting for burnup measurements is Nd148 which has a very similar fission yield in both U235 and Pu239 fission. Nd148 is normally determined by mass spectrometry with an isotopic dilution technique.

3) Plutonium determination.

Plutonium determination can be utilized for the measurement of Pu239 fissions or in a more general way for the measurement of the total burnup. In order to obtain the burnup data from plutonium measurements an accurate knowledge of the neutron energy distribution in the irradiated specimen is required. The plutonium measurement is the method which most depends on the knowledge of the neutron spectra.

4) Neutron flux monitoring.

Neutron flux monitoring at the surface of the irradiated specimen appears to be the most simple method of determining burnup because it allows one to avoid sampling the irradiated fuel and all of the radiochemical procedures required by the methods reported above. In order to determine the integrated flux received by the specimen, the ratio between the surface flux and the average flux in the specimen has to be calculated or experimentally determined.

The processing of the experimental data normally requires a knowledge of the neutron spectra. The data on the neutron spectra necessary to perform a good analysis of the experimental results are:

- a) Temperature of the Maxwellian distribution.
- b) Epithermal index r .
- c) Parameter β defined as the ratio of total to thermal + $\frac{1}{v}$ neutron captures in U238.
- d) Fast fission ratio R .

All the details on the utilization of these data are reported by Hart et al (1).

REFERENCES

- (1) R.G. Hart, M. Lounsbury, R.W. Jones, M.Y.F. Notley: Nucl. Sci. Eng., 18, 6 (1964).

1 - CALCULATION OF NEUTRON FLUXES AND RELATED PARAMETERS

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1.1 Introduction

The knowledge of the neutron energy distribution is important for the processing of the experimental data obtained by the different methods. Only the experimental data obtained by fission product determination allows one to calculate burnup values without requiring knowledge of the neutron spectra.

The calculation of neutron fluxes and related parameters have been made for the specimens to be irradiated in the DIRCE loop of the Ispra I reactor. The characteristics of the loop and the specimens have been changed after the calculations had been performed.

However the procedure of the calculation is always valid and can be utilized for the present conditions.

1.2 Problem Specifications

The geometric and physics specifications adopted for the capsule during the calculations are schematized in fig. 1.1.

We have considered two cases depending on the presence in the central region of UC or Pb.

In the case of UC (natural uranium) we have assumed an average temperature of 1000°C, the surface and center temperatures being 600°C and 1300°C respectively. The UC density was consequently assumed to be 13,6 gr/cm³.

On the contrary, room temperature has been assumed for the case of Pb. As for the organic liquid we have made the following assumptions:

HB fraction = 10%, temperature = 350°C, C/H = 1,29, density = 0,843.

The above described experiment is to be inserted at the center of the ISPRA-I reactor in the place of a normal fuel element: here it must be noted that the basic configuration of the reactor consists of 19 fuel elements inserted in heavy water disposed schematically as sketched in fig.1.2.

The fuel elements are in fact of square section, however the radius of the rods having an equivalent circular section is about 4,5 cm, which is the same as for the experiment to be inserted at the center.

Note that the distance from the reactor center to center of the first ring of elements is 16,3 cm. Since the height of the reactor is about 60 cm, while the height of the experiment is 15 cm, organic liquid is assumed to be above and below the experiment.

Four quantities have to be calculated, in the case of UC at the center of the experiment, and they are the following:

- 1) Westcott r inside uranium carbide. It is known that this parameter is correlated to the ratio between the epithermal and the thermal spectrum. In fact, in the Westcott conventions, the neutron density distribution per unit velocity interval can be written $\frac{n(v)}{n} = (1-f) \varphi_m(v) + f \varphi_e(v)$.

Where φ_m and φ_e are the normalized Maxwellian and epithermal density distributions, and f is the fraction of the total density in the epithermal distribution. The spectral index r is related to f by the formula $r = f \sqrt{\mu\pi}/4$, μKT being the cut-off energy for the epithermal distribution.

- 2) β inside uranium carbide. This parameter is related to U238 absorption, and is defined as the ratio between the total absorption rate and the thermal + $1/v$ epithermal absorption rate.
- 3) $\bar{\varphi}/\varphi_s$ related to uranium carbide. In fact, while $\bar{\varphi}$ represents the average flux in the UC element, φ_s represents the value of the flux at the external surface of the Aluminium cladding surrounding the UC element. Note that these fluxes have to be interpreted as integrated over the whole energy range.
- 4) R inside the uranium carbide. This parameter is defined as the ratio between the total number of fissions of both U235 and U238 over the whole energy range, to the total number of fissions of U235.

In the case of the presence of Pb instead of UC, only the first and third quantity, that is r and $\bar{\varphi}/\varphi_s$, can obviously be calculated.

It must be noted here that in the calculations we have performed we have assumed a zero derivative for the fluxes in the radial direction at a radius of 9.5 cm from the reactor center. This hypothesis is of course fairly valid when the experiment to be inserted at the reactor center is somewhat similar to the fuel elements of the reactor.

In the problem we have to study, this is not the case since the Ispra I fuel elements are quite different from the capsule mainly in the case of the presence of Pb. However, we have found that for the parameters we are searching for, which are related to the very internal part of the experiment, (i.e. the UC or Pb rod), the results do not vary when the zero derivative condition is moved from the radius of 9.5 cm to other nearby positions. As for the axial direction, we have assumed the dimensions of both the experiment and the reactor to be infinite. In fact, to take into account the height of the capsule would complicate every calculation probably without giving better results, since the height of the experiment is about 6 times its diameter.

1.3. Approximate calculations of the required parameters r , β , $\bar{\phi}/\phi_s$, and R in the UC rod.

1) Calculation of Westcott r .

In this approximate calculation of r we have first assumed that in UC the flux above $\sim 0,75$ ev has in fact a $1/E$ behaviour.

This approximation is probably rather good since the UC rod is surrounded by moderating materials and moreover contains carbon, which is a rather good moderator. The problem is then the determination of the spectrum in the very thermal region, that is from zero to $\sim 0,75$ ev. For this purpose we have utilized the code THERMOS, (1) developed at Brookhaven, which integrates the Boltzmann equation in the thermal energy region and in cylindrical geometry. Such a code assumes as a boundary condition with respect to energy an entering $1/E$ flux, and with respect to space a zero derivative condition at the external radius of the considered region. As we have said before this radius was assumed to be 9.5 cm.

A nuclear data library of 30 energy groups is incorporated into the code for the most used elements. The THERMOS code gives as output the neutron density as a function of space and energy; more specifically in each of the twenty space points chosen in the radial direction from zero to 9.5 cm, the neutron density is given in up to 30 energy

points from 0 to 0.75 ev. In such a way we get the spectrum from zero to 0.75 ev in the interior of the UC rod.

From the calculations we have performed we have seen that fortunately the $1/E$ shape just begins at the end of the thermal range, and this confirms the validity of our original hypothesis (apart from the possibility of flux fluctuations in the epithermal zone due to the presence of the U238 resonances).

In order to get the parameter r we calculate the following ratio Q :

$$Q = \frac{\int_0^{\infty} n(v) dv}{\int_{v^*}^{\infty} n(v) dv} \quad \text{where } v^* \text{ is a velocity at which the spectrum can be considered to have a } 1/E \text{ shape.}$$

Note that if v^* corresponds to the Cadmium cut-off energy, the above ratio is in fact known as the Cadmium ratio. We have, in our hypothesis of a $1/E$ flux for $v > v^*$,

$$Q = \frac{\int_0^{\infty} n(v) dv}{\int_{v^*}^{\infty} n(v) dv} = 1 + \frac{\int_0^{v^*} n(v) dv}{\int_{v^*}^{\infty} n(v) dv} = 1 + \frac{\int_0^{v^*} n(v) dv}{n(v^*) \cdot v^*}$$

Now, from the results of THERMOS we can easily calculate Q .

However, if we remember the Westcott formula,

$$\frac{n(v)}{n} = (1-f) \xi_m + f \xi_e \quad \text{we find:}$$

$$Q = \frac{1}{4r} \sqrt{\frac{\pi E(v^*)}{KT}}, \quad \text{where } T \text{ is the Maxwellian temperature.}$$

By comparing the two Q values we can easily find the Westcott r .

The value we obtain at the middle of the UC rod is $r=0,024$.

2) Calculation of β .

The value of β is correlated to the one of r by the following expression:

$$\beta = 1 + r \frac{I}{\sigma_{ao}} \sqrt{\frac{4T}{\pi T_0}} \quad \text{where } I \text{ is the resonance integral as calculated for the geometry considered, and } T \text{ is the neutron temperature.}$$

Since we know the values of r and T , the problem here is only to obtain the I value. The calculation of the resonance integral for a UC rod at a temperature of 1000°C can be done by means of nuclear codes such as ZUT TUZ ⁽²⁾(General Atomic) or ARES ⁽³⁾(Ato-

mics International). One can obtain the following expression: $I = A + B \sqrt{\frac{S}{M}}$ where $A = 4,12$, $B = 24,5$,

and S and M are the surface and mass of the UC rod respectively.

The value of I comes out to be 12,56b, and consequently we obtain $\beta = 1.198$.

3) Calculation of $\bar{\varphi}/\varphi_s$.

The estimate we have first made of this ratio is a very approximate one. In fact the method we have used is valid only with very restrictive hypotheses which in our case are not well verified. The hypotheses are that we have a rod of an absorbing material, which does not moderate, and which is not fissile, surrounded by a scattering and moderating material (having possibly an absorption cross section different from zero). In such a case there is a method and a corresponding code⁽⁴⁾ (THRILL developed at CETIS-ISPRA), which permits the determination of the ratio $\bar{\varphi}/\varphi_s$ at different energy intervals.

Clearly this is not the case, but we have obtained, using the THRILL code, an estimate of $\bar{\varphi}/\varphi_s$ for the case of a UC rod.

The values are the following:

Thermal region	= 0,90
Epithermal region	= 0,98
Fast region	= 0,95

4) Calculation of R .

Here too a rather approximate estimation has been done. However the precision required is not high, and we assume our method to be sufficiently accurate.

We have assumed the spectrum to be $1/E$ from 0,75 eV to 10 MeV and to be zero above this energy; this is certainly not the case since the flux generally rises above $1/E$ near 0.1 MeV, reaches a maximum at about 2 MeV, and then drops rapidly above 10 MeV. However from many calculations executed in quite similar cases we can assert that such fluctuations are not big ones.

Together with the assumption of the flux shape, we have assumed the σ_f of U238 to be zero everywhere except between 1,3 and 10 MeV, where a constant value of 0,6 b has been assumed.

As for the thermal part of the spectrum, we know it from the r calculation. From the relation

$$R = 1 + \frac{\int_0^{\infty} \sigma_{f38}(v) n(v) v dv}{\int_0^{\infty} \sigma_{f35}(v) n(v) v dv}$$

we obtain, at the center of the UC rod, a value of $R=1,013$.

1.4 Detailed Calculations of the parameters r , β , and $\bar{\varphi}/\varphi_s$.

Clearly, if we could have a good flux distribution in the whole energy range, without the hypothesis of a $1/E$ behaviour in the epithermal zone, the calculation of the required parameters would be much more reliable, and also much more straightforward. For this purpose we have thought of the possibility of using the THERFAST code,⁽⁵⁾ which is a modification of the THERMOS code developed at CETIS-ISPRA, permitting one to treat the range from 0 to 0.1 MeV.

The energy range has been divided into 30 groups. The first problem was the preparation of a good nuclear data library for the required elements for the 30 energy groups chosen.

We have utilized the code GGC⁽⁶⁾ (General Atomic) to obtain such a library. This code calculates, for a homogeneous region, the energy flux distribution corresponding to the given percentage of isotopes; then the code by means of an attached basic library of nuclear cross sections (specified at 200 energy groups), gives the flux averaged cross sections over the required broad groups.

In order to utilize GGC we had consequently to perform the homogenization of the given cell, that is, the experiment surrounded by a heavy water annulus. However, it is to be noted that mixing U238, having high resonance peaks, with other materials can lead to results very different from the ones corresponding to the real case, in which the U238 is all in a single central rod.

With the GGC code, one can fortunately correct the homogeneous calculations by taking into account the real geometrical configuration of U238. The self shielding correction is calculated by the code using the Nordheim method. It is interesting to compare the results one obtains by GGC for the absorber cross sections of U238 in the three different cases of dilution, 0 dilution, and a UC rod having the prescribed geometry and temperature. In fig. 1.3 the three curves are reported for the case of 24 energy groups, and one can note the strong difference between them. It is worthwhile to note the Los Alamos nuclear data library for the same number of groups, contains values comparable to the ones corresponding to the ∞ dilution case.

Once the nuclear data library was obtained and transformed into a form able to be read by the code THERFAST, we performed the spectral calculations as applied to the considered cell. A boundary condition of zero derivative for fluxes has been used at 9,5 cm.

The output of THERFAST consists of a matrix contain-

ning in each of the 20 space points, chosen from 0 to 9,5 cm, the neutron density in the thirty energy groups from 0 to 0.1 MeV. We have performed THERFAST calculations both in the case of UC and of Pb in the central zone.

Fig. 1.4 gives the spectrum as obtained at the middle point of the UC rod. Fig. 1.5 gives the neutron density and flux distribution in the different regions of the DIRCE loop.

Once having the fluxes at each point and for each energy interval, it was obviously possible to calculate the parameters r , β , and $\bar{\varphi}/\varphi_s$.

1) r Calculation.

As for r the method previously described was followed. Now we possess the entire spectrum and the Q value is calculated directly by means of the formula

$$Q = \frac{\int_0^{\infty} n(v) dv}{\int_{v^*}^{\infty} n(v) dv}$$

It is to be noted that the previously chosen value of v^* , that is 0,75, comes out to be a rather good one, since at that value of the energy the flux shows in fact a $\frac{1}{E}$ behaviour.

The value of r has been calculated at 4 points equally spaced inside UC rod.

The results are:

$$\begin{aligned} r &= 0,0246 \\ r &= 0,0244 \\ r &= 0,0239 \\ r &= 0,0229 \end{aligned}$$

2) Calculation of β .

Having the fluxes, the parameter β can be obtained independently and not through the r value just found. In fact, if we define X as being an element having the same thermal cross section as U238, but being $1/v$ in the epithermal region, then β can be found from the following relation

$$\beta = \frac{\int_0^{\infty} \sigma_{aU238}(v) v n(v) dv}{\int_0^{\infty} \sigma_{ax}(v) v n(v) dv}$$

By using the given absorption cross sections and the THERFAST fluxes we obtain, at the four points in UC, the following values:

$$\begin{aligned}\beta &= 1,205 \\ \beta &= 1,203 \\ \beta &= 1,198 \\ \beta &= 1,190\end{aligned}$$

3) Calculation of $\bar{\varphi} / \varphi_s$.

We can now write

$$\bar{\varphi} / \varphi_s = \frac{\int_{uc} dr \int_0^{\infty} n(r,v) v dv}{\left| \int_0^{\infty} n(r,v) v dv \right|_s}$$

By performing the above integrations, we get for UC rod $\bar{\varphi} / \varphi_s = 0,922$

We note that a satisfactory agreement has been found between the approximate and more detailed calculations, the one being a check for the other and, to some degree, viceversa.

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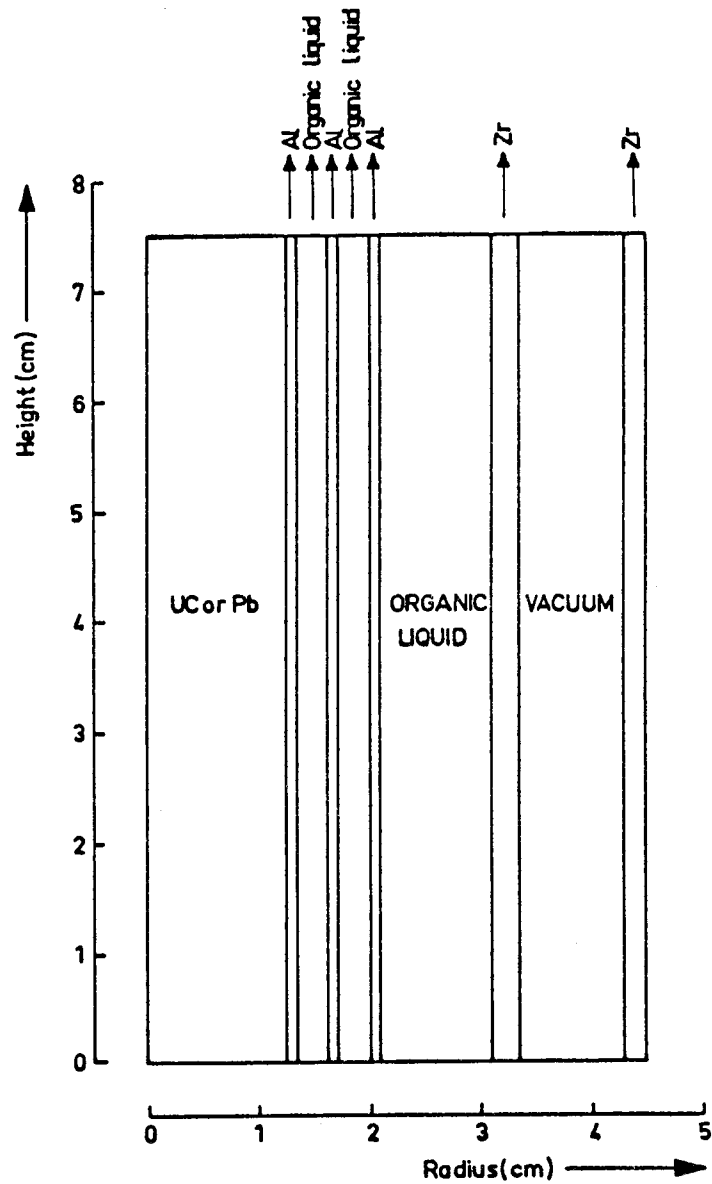


Fig. 1.1- UPPER RIGHT QUARTER OF THE CAPSULE

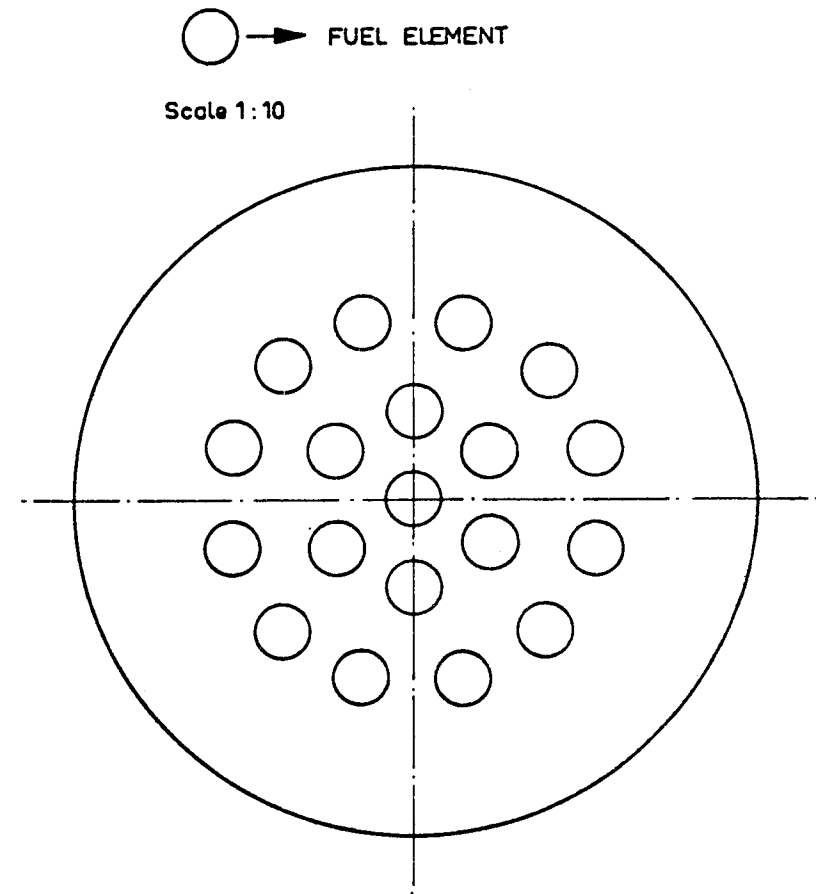


Fig. 1.2-SKETCH OF THE ISPRA I REACTOR CORE

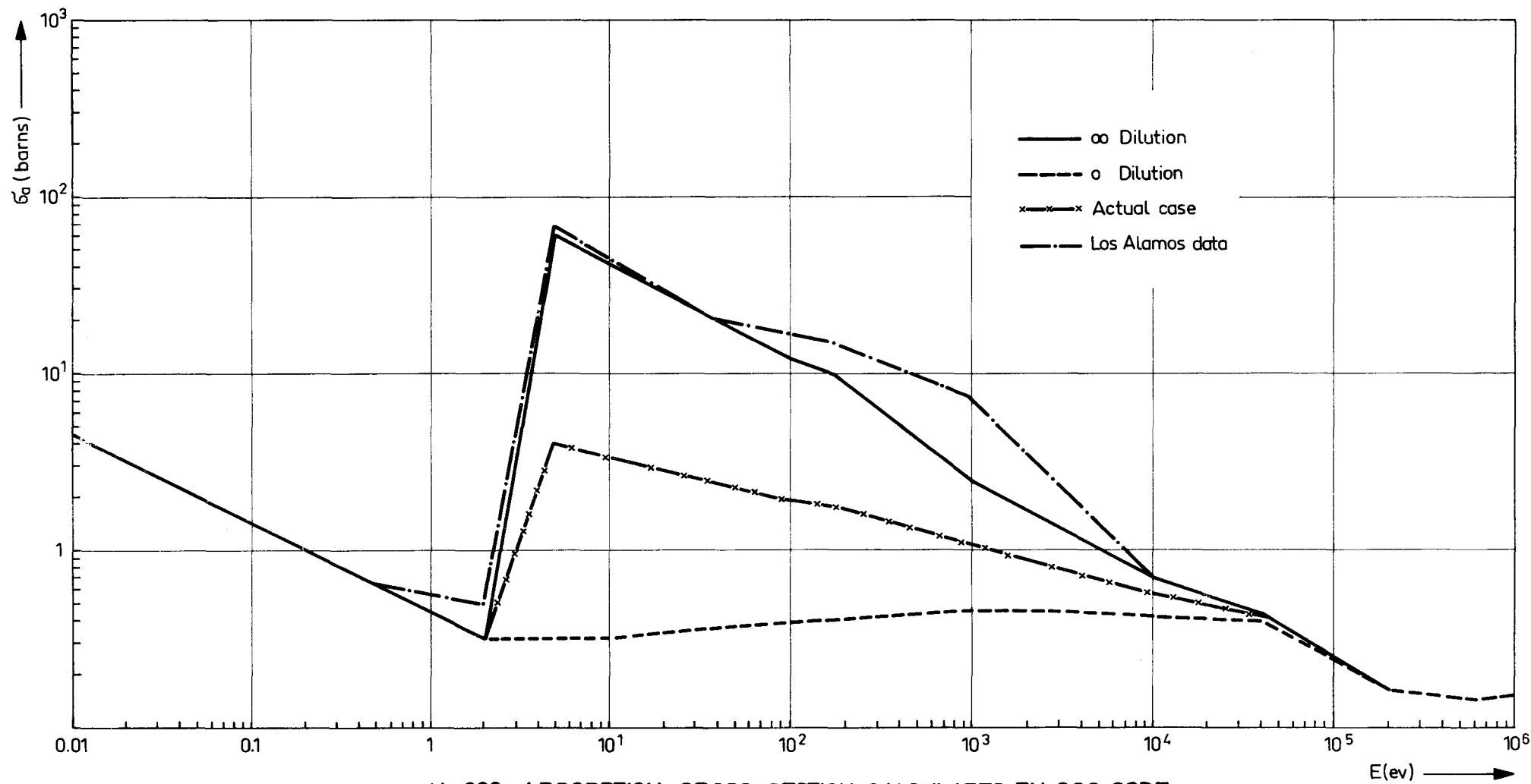


Fig. 1.3 U-238 ABSORPTION CROSS SECTION CALCULATED BY GGC CODE

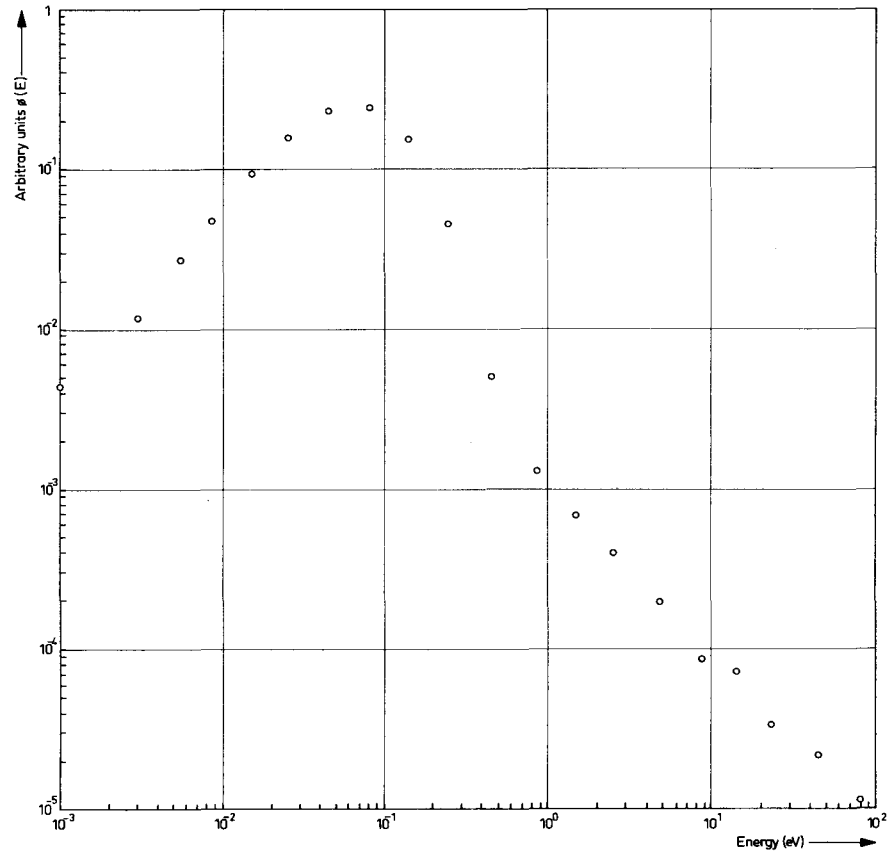


Fig. 1.4 CALCULATION OF NEUTRON SPECTRA BY TRANSPORT CODE-DIFFERENTIAL NEUTRON FLUX IN THE CENTER OF THE UC IN THE DIRCE LOOP

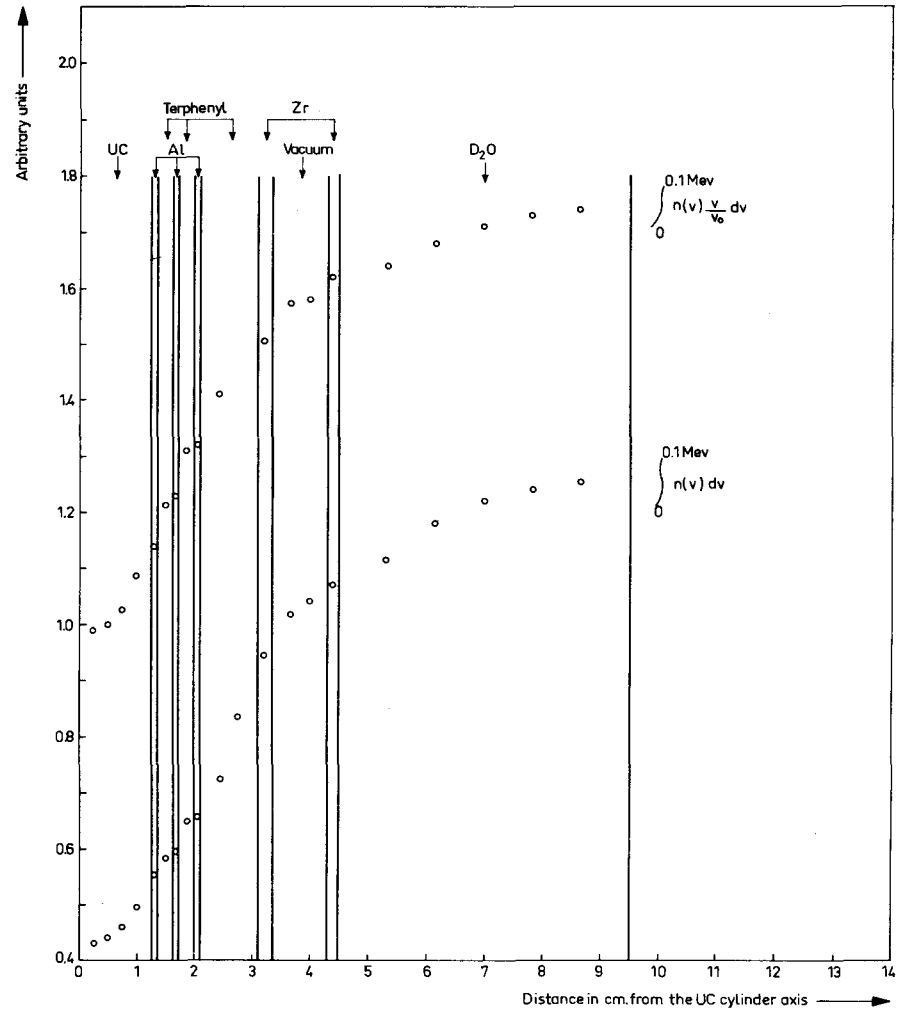


Fig. 1.5 CALCULATION OF NEUTRON DENSITIES AND FLUXES IN THE DIRCE LOOP BY TRANSPORT CODES

2 - DISSOLUTION OF UC SAMPLES

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G.Wagni - Nuclear Chemistry Laboratory
Chemistry Department.

2.1 Criteria in the choice of the UC samples for the measurement.

The burnup measurements of small samples of UC, taken from irradiated specimens, would greatly simplify the radiochemical operations. If the radioactivity of the samples is kept under a certain level the operations could be performed without utilizing heavy equipment such as a lead cell. Also all the problems connected with the transport of the samples and with the storage of the waste would be highly simplified.

However, operating with small samples it is possible to make large errors if such phenomena as diffusion of fission products and plutonium, and asymmetry of flux are present.

Radial distribution studies in UO₂ (1) have shown that cesium is grossly depleted in the high-temperature regions of the fuel and that plutonium may be depleted in regions with a high thermal gradient. Probably also other isotopes, which can be utilized in the burnup determination, diffuse in the fuel although in less important ways.

The asymmetry of flux is the other fact limiting the possibility of operating with small samples.

For these reasons, R.G.Hart et al (2), in burnup measurements of UO₂ specimens, have analysed only pellets corresponding to a complete cross section of the cylindrical specimens. Taking the complete cross section eliminated the necessity of integrating radially, leaving only the vertical integration.

The conditions for the specimens irradiated in the central position of the Ispra I reactor are quite favorable because flux symmetry can be assumed (however this symmetry will be experimentally checked).

Further the thermal conductivity is higher in UC than in UO₂ and the temperature gradients are consequently smaller. Moreover the diffusion coefficients in UC are normally smaller than the diffusion coefficients in UO₂.

The irradiations made by the Physical Chemistry Laboratory and planned by the Metallurgy Laboratory in DIRCE have periods of a few weeks. These relatively short periods will keep the diffusion processes at a low level.

For these reasons we hope to be able to obtain representative results from the UC irradiated by the Physical Chemistry Laboratory even operating with small samples removed by impact from the surface of the cylindrical specimens (See Section 7).

A more accurate sampling was not possible because the hot cell was not equipped for cutting the UC specimens.

The data obtained from these samples have to be modified on the basis of the radial and axial distribution of the fission rate in order to obtain the burnup data for the whole specimen.

Also in the DIRCE irradiations, which will be made under similar conditions, we expect to have diffusion phenomena of small importance and to be allowed to operate with small samples.

However, in the case that these predictions are wrong for the irradiations in the Ispra I core and mainly for the long period irradiations planned at Petten in conditions of flux asymmetry, we are equipping a lead cell with 10 cm thick walls for the dissolution of large amounts of UC - maximum 100 g - and for the sampling of small amounts of solutions for radiochemical determinations. For the samples irradiated at Petten, a limit to the amount of UC to be treated in the lead cell will probably be placed by the gamma activity level.

2.2 Methods for the dissolution of UC.

A complete review of the reactions of uranium monocarbide with water and with aqueous solutions of NaOH, HCl and H₂SO₄ is presented by M.J. Bradley et al (3). The data on the reaction of uranium monocarbide with HNO₃ are reported by L.M. Ferris and M.J. Bradley (4) and by J.R. Flannery et al (5).

We have selected data reported by these Authors for unirradiated and irradiated UC, prepared by arc melting of U+C by tungsten or graphite electrodes and with a carbon content 4.6 - 4.8%, because the characteristics of the UC to be utilized by the Metallurgy Laboratory are similar. Water (3).

Unirradiated uranium monocarbide reacts with water between 25° and 100°C yielding, as a hydrolysis product, a gelatinous, hydrous, tetravalent uranium oxide. The gas

evolved in the hydrolysis performed at 80°C (90 ml (STP) per gram of UC) consists of 88 vol.% methane, 9 vol. % hydrogen and 3 vol. % C₂ - to C₈ - hydrocarbons. 98% of the total carbon was present in the gaseous products.

The reactor irradiation of UC has an important effect on the hydrolysis. Two specimens irradiated to burnups of 0.6 and 1.6 at.% were inert to water at 80 and 100°C.

Another specimen irradiated at a burnup of 0.6 at.% hydrolyzed partially and the evolved gas consisted of a higher amount of hydrogen and lower amount of methane with respect to the hydrolysis of unirradiated UC.

Another specimen irradiated at a burnup level of 0.06 at.% hydrolyzed completely but the gaseous products also contained in this case more hydrogen and less methane than in the hydrolysis of unirradiated UC, and the carbon present in the gas was only 86% of the total carbon.

Sodium hydroxide (3).

The reaction of unirradiated UC with 6N NaOH at 80°C yields 104 ml (STP) of gas per gram of UC consisting of 25 vol.% hydrogen, 72 vol.% methane and 3 vol.% C₂ - to C₈-hydrocarbons. 93% of the total carbon was present in the gaseous products.

A specimen irradiated at a burnup level of 0.6 at.% was inert to 6N NaOH at 80°C. Gas evolution from a 0.06 at.% burnup specimen was considerably slower than from the unirradiated carbide, although the reaction did go to completion.

The reaction of this specimen produced more hydrogen and less methane than the unirradiated carbide. Only 60% of the original carbide carbon was found in the gas from the irradiated specimen.

Hydrochloric Acid (3).

Both unirradiated and irradiated UC reacted readily at 80°C with hydrochloric acid solutions. In general, the irradiated specimens were more reactive than the unirradiated. In the hydrochloric acid reaction, the irradiated carbides again yielded more hydrogen and less methane than unirradiated monocarbide; however the irradiation effect was less pronounced than that found with water or NaOH. Eighty to ninety % of the original carbon in the irradiated carbides was found in the gaseous products, while the uranium product was a solution of UCl₄. Although the HCl concentration did effect the rate of reaction, the gas composition was about the same with both 2 and 6 M acid.

Sulfuric Acid (3).

The reactor irradiated specimens also reacted with 6M H₂SO₄ at 80°C. Again the amount of hydrogen produced

increased with increasing burnup, while the amounts of methane and carbon recovered in the gas decreased. The uranium product was the insoluble $U(SO_4)_2 \cdot 4 H_2O$.

Nitric acid (4) (5).

Unirradiated and irradiated uranium carbide dissolved in 2 to 16 M HNO_3 yielding gases comprised mainly of NO_2 , NO and CO_2 . The off-gas composition varied with the acid concentration. Traces of N_2O were found, but no CO , H_2 , or gaseous hydrocarbons were detected. The carbides were almost completely passive in very dilute (0.001 to 0.5 M) acid. Between 30 and 50% of the carbide carbon was converted to species soluble in nitric acid irrespective of the carbide, reaction time, or variations of acid concentration between 2 and 16 M.

The soluble organic compounds mainly consist of oxalic acid (0 to 11 % of the carbide carbon), mellitic acid (2 to 9% of the carbide carbon) and other unidentified compounds which are probably aromatics highly substituted with - COOH and - OH groups. (6)

These organic impurities may interfere in the recovery of uranium and plutonium by solvent extraction. A partial degradation of the mixture of organic impurities to carbon dioxide can be obtained by strong oxidation.

2.3 Experimental results on the UC dissolution.

The uranium carbide to be utilized in the irradiations of the Metallurgy Laboratory is prepared by NUKEM. (Nukem, Nuklear - Chemie & Metallurgie G.M.B.H., Wolfgang 645 Hanau, Main, Germany).

The preparation process is based on the reaction $UO_2 + 3C \rightarrow UC + 2 CO$ at $1700^\circ - 2000^\circ C$ and uses arc melting of UC with graphite electrodes. The UC density is 13.6 g/cm^3 .

The stoichiometric content of carbon in uranium monocarbide is 4.80%. The analysis has shown a content of $C+O+N = 4.75 \pm 0.2\%$. The content of $O + N$ in the first preparations was $\sim 0.15\%$ and in the most recent preparations $\sim 0.05\%$. The UC_2 amount was very small; small amounts of elemental uranium may be present.

Experiments have been done in our laboratory on the dissolution of UC samples prepared by NUKEM.

It is convenient for separations of uranium and plutonium by solvent extraction to use a nitric acid solution. In order to avoid the formation of organic compounds in the direct attack with HNO_3 it would be useful to perform a preliminary hydrolysis of the UC.

We have hydrolysed three samples, 82 g, 52 g, and 41 g with water at $90^\circ C$ corresponding to complete cross sections of a cylinder 25 mm in diameter; after 24 hours

the hydrolysis was not yet completed. The dissolution of the samples was obtained by addition of conc HNO_3 . In order to reduce the dissolution time we have decided to operate directly with HNO_3 and to investigate the interference of the organic compounds on the extraction processes (see Section 6). It has been found that the organic compounds don't interfere in the quantitative Uranium extraction by TBP. In the extraction of Plutonium a quantitative yield is not required and eventual interferences reducing the yield are not troublesome. A direct treatment of the UC with 2M HNO_3 has permitted complete dissolution of a 50 g sample in 14 hours, operating at 90°C.

Also the direct attack on UC by HCl and H_2SO_4 has been tested. The dissolution of a 40 g sample has been obtained in about 3 hours operating with conc HCl. Small samples have been dissolved in a few hours in 20% H_2SO_4 .

The UC irradiated by the Physical Chemistry Laboratory is arc melted and has a carbon content of $5 \pm 0.1\%$, which is higher than the stoichiometric content - 4.8%. Experiments made with HCl and HNO_3 on unirradiated UC have shown that it dissolves in a way similar to the NUKEM product.

The dissolution of the irradiated samples required a longer time with both 3M HCl and 3-6M HNO_3 .

A short investigation has been performed on the possibility of preventing the polymerization processes, producing the organic compounds in the direct attack on UC by HNO_3 , by addition of nitrobenzene or benzophenone. Experiments have been performed using small samples (~ 1 g) of UC and 3M, 6M and 9M HNO_3 . No difference has been observed with and without nitrobenzene and benzophenone. However we believe that a more accurate investigation could give interesting results.

In conclusion in the future we plan to utilize dissolution by HNO_3 , using different molarities depending by the amount of UC to be dissolved. The dissolution of high activity samples will be performed in a lead cell. In the lead cell the sampling of small aliquots from the bulk will be made. The activity of these aliquots will allow one to operate in a glove box or a fume hood without special shielding. The lead cell to be utilized has 10 cm thick walls and is equipped with Model 7 master slave manipulators and with a glove box for handling α activities.

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3 - THE ISOTOPIC DETERMINATION OF U-235 AT LOW
CONCENTRATION BY OPTICAL SPECTROGRAPHY.

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A paper has already been published (1) on the spectrographic determination of Uranium isotopic ratios for low contents of U235. The operating conditions, using a Jarrell-Ash Mark IV, 3.4 m Ebert spectrograph, were the following:
grating: 7.500 grooves/inch blazed at 33° and 61°
grating setting: 61° equivalent to λ 4244 14th order
slit width: 10 μ

Filter: Kodak wratten 2 A

Excitation: D.C. Arc 10 A

Pre-burn period: 6 secs

Exposure period: 30 secs

Electrodes: National SPK grade

Arc gap: 4 mm

Samples: 20-50 mg in the form of U₃O₈

Plates: Kodak S.A. 1

Emulsion calibration: Iron arc through 7 step filter

Precision studies, made with the above described procedure, were carried out at the natural uranium level by arcing twenty-one samples. A standard deviation of 0,0398, with a σ % = 5,62 was obtained.

Although this problem can be solved by optical spectroscopy with good accuracy, the error coming from the plate and the microphotometer can play a very important role. On the other hand, a D.C. arc discharge, very luminous and highly sensitive, shows a markable instability associated with a continuum background and a Doppler broadening of spectral lines, which is impractical when high precision is required. Moreover a 20 - 50 mgr sample is needed for a single determination.

The necessity of overcoming these difficulties has led us to study the hollow cathode lamp as an alternative source of excitation. This source, widely used in the study of isotopic spectra, shows high stability, lack of spectral background, and highly reduced Doppler effect; however its lumi-

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nosity is poor. These properties allowed the determination of U235 down to the naturally occurring level by direct reading methods, and of enriched Uranium by photographic procedures.

The working currents ranged between 80 and 300 mA.

In order to increase the luminous energy of the discharge and consequently to record photographically the low U235 concentration, lamps working at high energy rate were investigated. For this purpose, the lamp requires an efficient cooling system to get a sharp line definition, a suitable geometry to avoid the tendency of the discharge to turn in the arc condition and a good electrical isolation between the anode and the cathode body to concentrate the negative glow into the cathode. Since a commercial lamp working at high current gave no satisfactory results, several models of lamps were built and tested in our laboratory.

The highest excitation of the Uranium is obtained when the cathode is made from the Uranium metal. However the practical impossibility of working under these conditions has led us to study ways by which the uranium sample could be deposited as an adherent layer in the bottom of a suitable cathode. As cathodic material, stainless steel, nickel and graphite were investigated, the Uranium being introduced either as uranyl nitrate, or as a refractory oxide, or as a refractory oxide dissolved in hydrofluoric acid.

The first two compounds were discarded because of the discharge instability and of the poor luminosity.

The layer obtained the third way allows a stable discharge condition lasting about 60' to be reached after 15' warm-up. However with a stainless steel cathode a preferential excitation of Cr was observed, the Uranium being excited only to low extent and not affected by large variations in the discharge parameters (type and pressure of the inert gases). A slight increase of the uranium intensity was obtained using nickel cathodes.

Graphite was finally chosen due to its high sublimation temperature and excitation potential.

Moreover its spectrum is free from lines and background.

On the other hand the graphite absorbs gases and a considerable quantity of the solution introduced in the cathode, causing a lower sputtering efficiency in the glow.

To overcome the diffusion of the solution into the graphite, the cathodes were first dipped in a solution of paraffin in CCl_4 and the excess of paraffin was evaporated.

Fig. 3.1a shows the microphotometer tracing obtained by D.C. arc excitation.

Fig. 3.1b, 3.1c, 3.1d, show the microphotometer tracing obtained with the stainless steel hollow cathode and U metal, with nickel hollow cathode and $U_3O_8 + HF$, and with graphite hollow cathode and $U_3O_8 + HF$ respectively. We are now performing studies on the reproductibility and the precision which can be obtained with the source.

In the near future we intend to study the application of a direct reading system with mechanical displacement of photomultipliers along the entire spectral region in order to achieve the maximum refinement and improvement of the technique.

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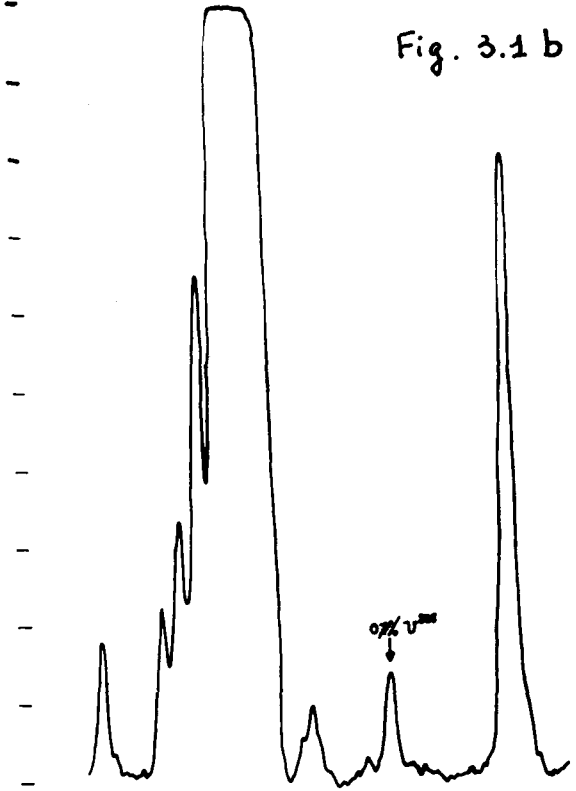
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Isotopic analysis of U by an optical spectral method.
Part I, Energia Nucleare Vol. 12 n° 6 Giugno 1965.

D.C. ARC 10 A U_3O_8

STAINLESS STEEL HOLLOW CATHODE
U METAL (250mA)

Fig. 3.1 a

Fig. 3.1 b



NICKLE HOLLOW CATHODE
 U_3O_8+HF (500mA)

GRAPHITE HOLLOW CATHODE WITHOUT
PARAFFIN U_3O_8+HF (500 mA)

Fig. 3.1 c

Fig. 3.1 d

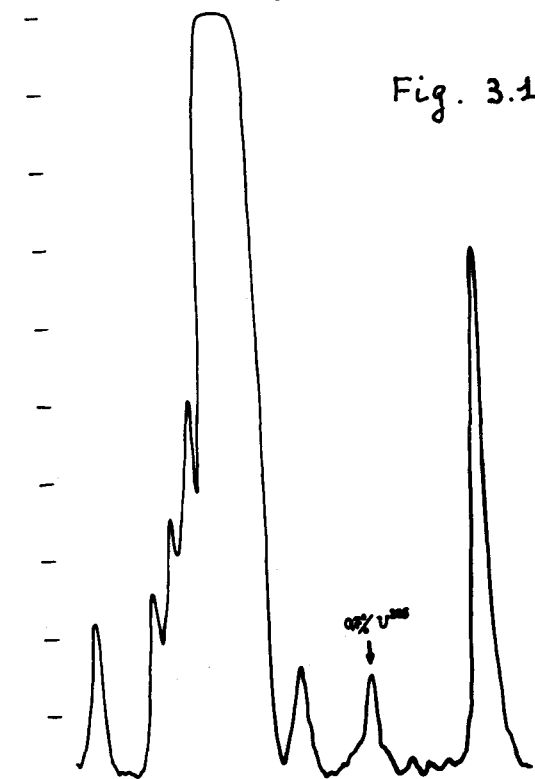
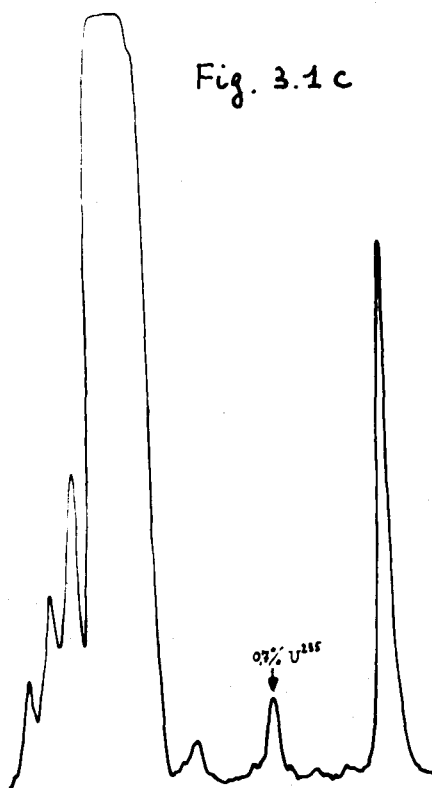


Fig. 3.1 MICROPHOTOMETER RECORDINGS
OF NATURAL URANIUM

4 - MEASUREMENT OF U235 CONTENT IN URANIUM

BY ACTIVATION TECHNIQUES.

M. Bresesti

Nuclear Chemistry Laboratory
Chemistry Department

4.1 Introduction.

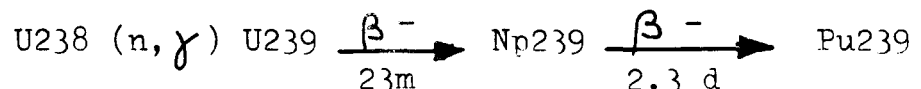
The activation techniques have been utilized in the determination of the U235 of depleted uranium, performed in collaboration by F. Rustichelli and G. Maracci of the Experimental Neutron Physics Laboratory - Reactor Physics Department - and M. Bresesti of the Nuclear Chemistry Laboratory - Chemistry Department. In these measurements we have obtained information on the accuracy that can be obtained by activation techniques and on the possibility of utilizing these techniques for the burnup determinations. The measurements reported here have been made by two different activation techniques in the two laboratories. For natural uranium, taken as a reference, we have assumed a U235 content of 0.712% by weight.

4.2 Experimental.

Method 1.

Pairs of 1/10 mm thick natural Uranium and 2/10 mm thick depleted Uranium foils, 12 mm in diameter, have been irradiated in the thermal column of the Ispra I Reactor in a thermal flux of about 10^9 n/cm²sec. The total gamma activity of the fission products has been measured after a few hours cooling using a 1 $\frac{3}{4}$ " x 2"

NaI(Tl) scintillation spectrometer with a threshold set at 0.72 MeV, as suggested by J.R. Wolberg et al. (1). The capture reaction in U238 and the decay chain of the resulting U239 nuclide is the following,



The maximum energy of the betas and associated brems-

strahlung is 1.2 MeV for U239 and 0.72 MeV for Np239. The gammas associated with these beta decays have lower energies than the maximum beta energies. Therefore, after a cooling period of a few hours to allow the U239 to decay, the fission product gamma activity can be measured free from capture product interference with a threshold set at 0.72 MeV.

The depleted and natural Uranium foils have been alternately measured and the ratio between the gamma activities has been found to be constant with time. The gamma activity per mg of Uranium has been assumed to be proportional to the U235 content, the fission of U238 being negligible under the flux conditions present in the thermal column. The natural activity of the foils has been subtracted, and small corrections for neutron self-shielding (2), for fission product losses due to the recoils (3) and for dead time losses in the counter have been introduced. The total correction is about 2%. The results are reported in table 4.1.

Method 2.

Pairs of natural Uranium and depleted Uranium samples have been irradiated in the graphite reflector of the Ispra I Reactor, close to the heavy water tank, utilizing a thermal flux of 2×10^{13} n/cm²-sec which is much higher than in the thermal column. The Uranium samples were wrapped in Al foils, which were thick enough to stop all the fission products. After irradiation both the Uranium and Aluminium were dissolved in 6N HCl and sources were prepared using suitable amounts of solution in order to have similar gamma activities in both the natural Uranium and depleted Uranium sources.

The 1.60 MeV gamma ray photopeak of La140, daughter of the fission product Ba140 (12.8 d half life), has been evaluated using a 3" x 3" NaI(Tl) crystal connected to a multichannel pulse height analyser. This gamma is emitted with an intensity of $96.1 \pm 0.8\%$ (4). The photopeak can be evaluated without interference from other gamma emitters present in the mixture of fission products, starting from about ten days after the end of the irradiation and continuing for a period of a few months (5). By following the decay of La140, which reaches transient equilibrium with Ba140 in about 2 weeks, it is possible to check for the absence of gamma activity in the 1.60 MeV region due to capture products from Uranium impurities. The photopeak activity per mg of Uranium has been assumed to be proportional to the U235 content. Small corrections have been introduced for neutron self-shielding (2) and for the U238 fission contribution which has been determined by irradiation

tion under Cadmium. The total correction is 1.6%. Results are reported in Table 4.1.

The results of these activation techniques have been checked using a double fission chamber placed in the thermal column of the Ispra I Reactor. In this counter, shown in fig. 4.1, the fission rates of natural and depleted Uranium, deposited on Platinum discs by vacuum evaporation, have been measured. The areal density of Uranium was about 0.20 mg/cm² and had previously been determined in each target by an X-ray technique in the Analytical Laboratory of the Chemistry Department.

The chamber was filled with Argon containing 2% Nitrogen to a pressure of 1 atm. The anode voltage was 500 V and the slope of the discriminator curve, expressed as $\frac{\Delta \text{ counts}}{\text{total counts}}$

/mv was 2.1×10^{-5} .

Background was measured by operating with a Uranium deposit in one chamber to monitor the flux and with a Pt disc without Uranium deposit in the other chamber. The background was found to be proportional to the neutron flux.

The measured fission rates have been corrected for dead time and fission product absorption (3) in the Uranium deposits. The fission rate per mg of Uranium has been assumed to be proportional to the U235 content, the fast fission of U238 being negligible in the flux conditions of the thermal column.

The results are reported in Table 4.1.

A measurement by the BCMN laboratory at Geel, using mass spectrometry gave a U235 content of 354 ± 14 ppm (95% confidence) for the same sample, in good agreement with the activation techniques.

4.3 Conclusions.

The observation of the agreement of the data obtained with the different methods allows one to conclude that the activation techniques can be utilized in the burnup determination when an accuracy of about 2% is sufficient.

Naturally the accuracy required depends on the depletion of U235. The accuracy of 2% is not good for burnup determinations in DIRCE irradiations where a maximum U235 depletion of 10% is expected and on the contrary can be sufficient for the irradiations at Petten where much higher U235 depletion is expected. If the activation techniques are to be utilized, the uranium to be irradiated for the U235 content determination has to be carefully

purified from the fission products and transformed into a chemical compound of known uranium content. In fact, a known amount of uranium has to be irradiated.

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Table 4.1 - U235 content in depleted Uranium measured by different methods.

Method	Experiment	U235 content (ppm by weight)	Average value (95% confidence level) (6).
Total gamma counting with a 0.72 MeV threshold	1	356	355 ± 4
	2	359	
	3	354	
	4	357	
	5	351	
La140 1.60MeV gamma photopeak measurement	1	355	358 ± 9
	2	361	
	3	345	
	4	350	
	5	370	
	6	370	
	7	356	
Fission rate measurements by a double fission chamber	1	357	357 ± 10
	2	353	
	3	361	

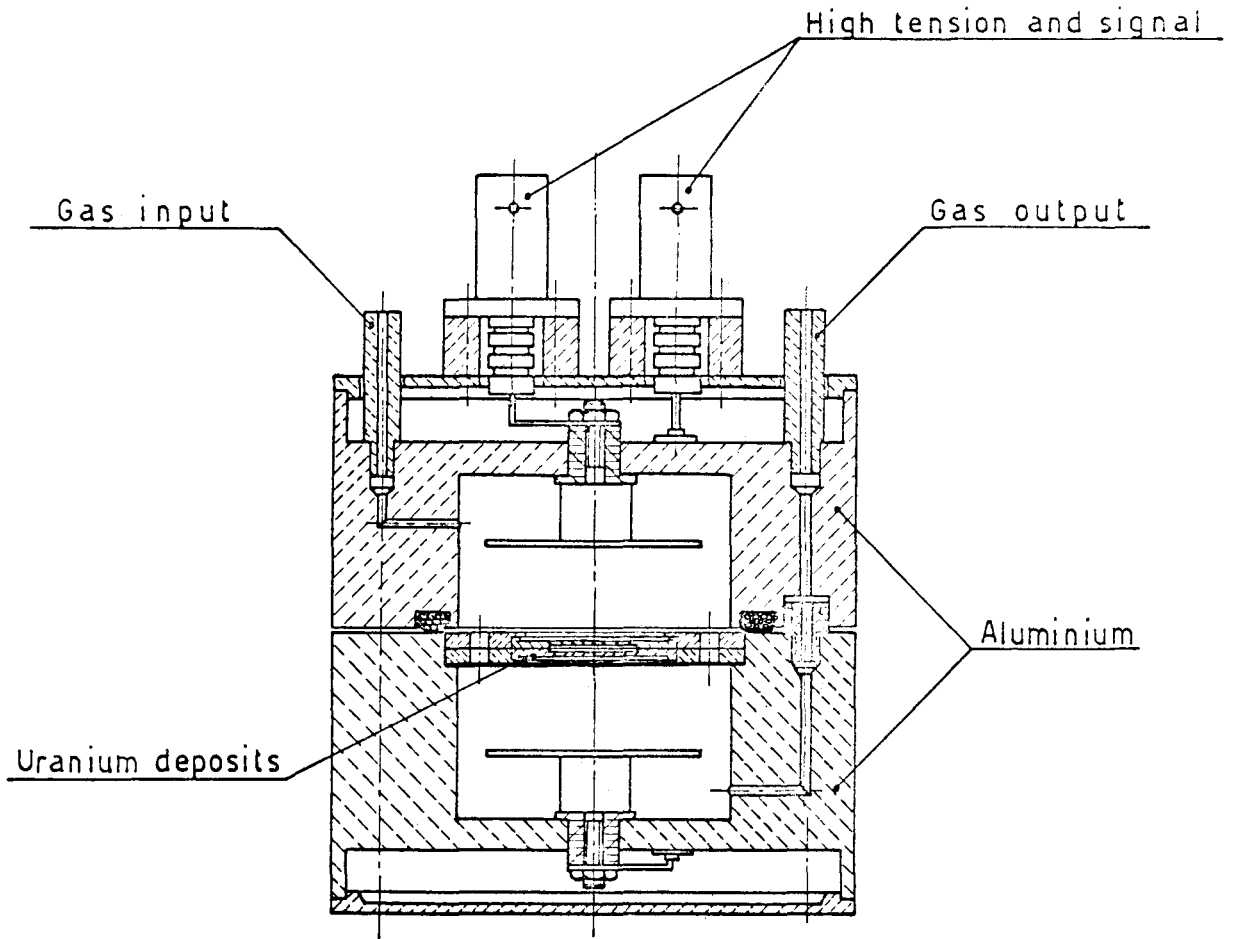


Fig. 4.1 - DOUBLE FISSION CHAMBER

5 - DETERMINATION OF TOTAL URANIUM IN PURIFIED SOLUTIONS
FROM THE DISSOLUTION OF IRRADIATED UC.

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Chemistry Department

For this determination the following methods have been taken into consideration and studied in some detail:

- 5.1 - Extraction of uranium with trioctylphosphineoxide, and colorimetric determination with dibenzoylmethane.
- 5.2 - Coulometric determination at controlled potential.
- 5.3 - Other methods.

5.1 Colorimetric determination with dibenzoylmethane. (1) (2)

Principle of the method.

The uranium is separated from the aqueous solution, and from interfering elements contained therein, by extraction with trioctylphosphineoxide (TOPO) dissolved in cyclohexane. The aqueous solution of uranium may contain sulfuric, nitric or perchloric acid; chloride ions interfere. The colorimetric determination is based on the formation of a yellow complex between uranium and dibenzoylmethane in the organic phase. Na, K, Ca, Cd, Co, Cr⁺³, Al, Y, La and Fe⁺² do not interfere if present in less than a 300 fold excess of the uranium content. Cr⁺⁶, V, Fe⁺³, Th, Zr, Ti and phosphate ions do interfere in the determination; they may either be reduced to a lower valency state or complexed by suitable additions of masking agents. (2).

Experimental results.

The minimum quantity of uranium which can be determined by this method is about 20 μg . The optimum concentrations are between 200 and 2000 μg . Starting from a uranium standard solution the following precisions have been obtained experimentally:

Amount of uranium determined μg	Coefficient of variation % = $\frac{S \cdot 100}{x}$
100	1.18
300	0.64
500	0.80

These values have been obtained from a series of 10 determinations for each concentration level. 10 determinations per day can be performed by one person, if the activity level of the solutions obtained for the analysis allows this to be done in a normal chemical laboratory.

5.2 Coulometric determination at controlled potential.

Principle of the method.

The solution to be analyzed must be pure and contain the uranium in the hexavalent state in a sulfuric acid medium. Nitric and hydrochloric acid solutions can be easily transformed into sulfate solutions by evaporation with concentrated sulfuric acid. The method is based upon the reduction of U (VI) to U(IV) at a potential of - 0.450 V/SCE. If the determination is performed in 0.1N oxalic acid (3) traces of iron and phosphate don't interfere.

Experimental results.

A coulometer of the type O.R.N.L. Q - 2005 x 50 combined with a digital voltmeter has been utilized. Starting from a standard solution in a sulfuric acid medium the following values have been obtained:

Amount of uranium determined μg	Coefficient of variation % = $\frac{6.100}{x}$
4000	0.35
700	0.30
350	0.88
70	1.27

These values have been calculated from a series of 10 determinations for each concentration level.

The method of determination for samples containing 4000 and 700 μg of U has been developed for use as a routine analysis. With this method the "automatic stop" feature of the coulometer is used. More precise results can be obtained when the method of subtracting the residual current is applied.

Experiments are under way in order to study the coulometric determination in media different from sulphuric acid (3) (4) (5) and to obtain still better reproducibility of the determination. An installation, allowing the determination in moderately radioactive solutions has been ordered.

5.3 Other methods.

In order to check, if necessary, the yield of the different steps of the purification of uranium from fission products by solvent extraction, to be performed by the Nuclear Chemistry Laboratory, the following analytical tech-

niques can also be utilized:

- 1) For high uranium concentrations: gravimetry, volumetry and colorimetry with H_2O_2 .
- 2) For trace amounts of uranium : fluorimetry.

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6 - URANIUM PURIFICATION FROM FISSION PRODUCTS

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6.1 Introduction.

The Uranium samples to be analyzed for Uranium content (see Section 5) and for isotopic composition by Optical Spectrometry (see Section 3) have to be purified in order to reduce their activity to the levels permitted in the Analytical Chemistry Laboratory.

When the samples are analyzed for Uranium content, the purified Uranium has to be recovered quantitatively. For the samples used for isotopic analysis the recovery of Uranium does not have to be quantitative.

A particularly good decontamination will be required if the neutron activation method (see Section 4) is utilized for U235 content determination.

6.2 Experimental.

For the separation of Uranium from fission products the TBP - hexane extraction has been tested. The yield of the Uranium extraction has been measured for solutions of unirradiated UC dissolved in nitric acid. Decontamination factors have been measured for solutions of irradiated UC dissolved in nitric acid.

Reagents.

Extraction reagent. Dissolve 56 g of $\text{Al}(\text{NO}_3)_3 \cdot 9 \text{H}_2\text{O}$ and 5 g of citric acid in 100 ml of 1N HNO_3 . Mix 2 volumes of this solution with 1 volume of a 6.7 M Li NO_3 solution.

Lithium nitrate solution, 6.7 M. Dissolve 231 g of Li NO_3 in water and dilute to 500 ml with water.

TBP - hexane reagent. Dissolve 30 volumes of TBP in n - hexane and dilute to 100 volumes with n-hexane.

Procedure.

Pipet in a centrifuge tube 5 ml of TBP solution, 4 ml of extraction reagent and 500-1000 μl of the Uranium solution. Close the tube, stir by hand for 3', centrifuge the two phases for 3' and separate the organic phase.

Wash the organic phase twice with 3N HNO₃.
Strip Uranium from the organic phase using 0.01 N
nitric acid.

6.3 Results.

Three experiments have been performed on Uranium solutions (50-100 g/l) obtained by dissolving unirradiated UC in nitric acid of different molarities. The amount of Uranium separated was between 20 and 100 mg. The yield of the separation was measured by the Analytical Chemistry Laboratory by determining the Uranium content in the aqueous phase after the extraction.

The yield of the separation in the three experiments was found to be about 99.9%. We have to point out that this yield was obtained in the presence of the organic compounds formed in the nitric acid dissolution of UC. The organic compounds appear not to interfere. The decontamination factors, determined by γ -spectrometry measurements on the initial solution of irradiated UC and on the final 0.01N nitric acid solution, are about 5×10^4 for Ce144 and Cs137 and about 10^3 for Ru106 and Zr95 - Nb95.

7 - FISSION PRODUCT DETERMINATION BY GAMMA SPECTROSCOPY.

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7.1 Introduction.

When the number of atoms of a nuclide formed in fission is known it is possible to determine directly the number of fissions by utilizing the fission yield of the nuclide. The calculation can be performed without taking into account the neutron energy distribution in the irradiations.

The most simple method of measurement of radioactive nuclides is gamma spectroscopy, which in many cases allows one to operate without chemical separations. However, to obtain the number of atoms of a radioactive nuclide formed in fission from activity measurements it is necessary to know the decay constant of the isotope and the history of the irradiation in order to calculate the decay correction. When the half life of the radioactive nuclide is not long in comparison with the irradiation time, the decay correction is important and a poor knowledge of the decay constant or irradiation history can introduce strong errors. Other sources of inaccuracy in the burnup determination by radioactive fission product measurements may be the poor knowledge of the decay scheme and of the neutron capture cross section of the nuclide. Also, an error in the knowledge of the fission yield is directly transmitted to the determined value of burnup. The situation is more complicated when the nuclide derives from the fission of two or more isotopes, for instance U235 and Pu239 in the irradiation of Uranium.

Cs137 is an isotope particularly interesting for its long half life (30 years), and high fission yield which is similar for U235 and Pu239 fission. The half life, the decay scheme and the fission yield of Cs137 are reasonably well known, and its measurement by γ spectrometry does not present particular difficulties. However Cs137 can diffuse inside high temperature fuels(1) and also fix in the cladding (2). The high tendency to diffuse can be explained by the presence of elementary cesium in a gaseous state at temperatures higher than 670°C. The diffusion phenomena depends on the temperature, the temperature gradient, the structural characteristics and the irradiation period of the fuel. A calculation of the importance of the diffusion phenomena as a function of the irradiation conditions is not easy and when the utilization of

the Cs137 analysis seems to be convenient, the best procedure is to perform a preliminary comparison of the burnup data obtained by a Cs137 determination with the data obtained by the determination of other fission products having a smaller tendency to diffuse, or by different methods.

The diffusion of other elements such as alkaline earths and rare earths is certainly smaller. In these groups the fission products Sr90 (28 y. half life) and Ce144 (284 d. half life) seem to be particularly interesting. However Sr90 is a pure β^- emitter and the daughter Y90 (64.5h half life), in equilibrium with the father, presents a very weak gamma intensity and its determination would require purification from other fission products before the activity measurement. Therefore our interest has been particularly fixed on the gamma emitting Ce144 which is in equilibrium with the daughter Pr144 (17 m. half life) as an isotope to be measured together with Cs137.

Another isotope of interest is Ru106 (365 d half life) in equilibrium with the daughter Rh106 (30s half life), which presents a large difference between the fission yields for U235 and Pu239. This difference can allow a determination of the single contributions of U235 and Pu239 to the total fissions as shown in the following pages of this Section.

For short irradiations, and burnup analyses to be performed in a short time, we have considered in some detail the possibility of measuring La140 which is in equilibrium with the fission product Ba140 (12.8 d. half life). For short irradiations, the measurement of the pair Zr-95 (65 d half life) - Nb95 (35 d half life) also appears to be convenient.

In this Section the most important observations obtained in the analysis of fission product mixtures by γ spectrometry with NaI(Tl) and Germanium Li-drifted detectors, are presented. Also the first results obtained in the burnup determination on UC samples irradiated by the Physical Chemistry Laboratory are reported.

7.2 Gamma spectroscopy by NaI(Tl) crystal.

The gamma spectrometer was formed by a 3" x 3" NaI(Tl) crystal (Harshaw integral line) connected with a multichannel pulse height analyzer.

In previous experiments we had observed the possibility of measuring the 1.60 MeV γ -photopeak of La140 directly from the mixture of fission products, and this measurement was utilized also in the determination of the U235 content in depleted Uranium (Section 4). In the present experiment we have determined how long after a short time irradiation

of Uranium the 1.60 MeV γ -photopeak of La140 was free from interference from other fission product gamma rays. La140 (40.2 h. half life) reaches transient equilibrium with the parent Ba140 (12.8 d. half life) in about 2 weeks. The fission yield of Ba140 in the fission of U235 is 6.44% (3), and in the fission of Pu239 is 5.68 % (3), the independent fission yield of La140 being negligible. The 1.60 MeV gamma is emitted with an intensity of $96.1 \pm 0.8\%$ (4).

The decay curve of the 1.60 MeV gamma - photopeak (figure 7.1) has been obtained from a sample of uranium carbide irradiated for 4 hours in the Ispra I reactor in a thermal flux of 2×10^{13} n/cm²sec. The decay curve is a straight line for about 100 days after the end of the irradiation and the half life determined from its slope is in agreement with the 12.8 day value reported in the literature. For decay times longer than 100 days the decay curve appears to deviate from the straight line probably due to the interference of the gammas of Pr144. However, by suitable corrections the γ -photopeak of La140 could be evaluated also under these conditions. In figure 7.2 the gamma spectrum of the fission product mixture about 1 month after the end of the irradiation is presented.

In fission product mixtures to be measured after a long decay period, the Pr144 2.18 MeV γ -photopeak appears to be free from interference of other fission product gammas. The interfering element for a few months after the end of the irradiation is La140 which presents gammas of 2.37 (0.86%), 2.53 (3.0%), 2.89 (0.082%), and 3.10 (0.035 %) MeV (4). Interference can also derivate from Rh106, (30 s. half life), daughter of the fission product Ru106 (365 d half life), which presents several gammas of low intensity in the energy range between 2 and 3 MeV. However, the Rh106 interference is important only for fuels irradiated in such a way as to have a high contribution of Pu239 fissions to the total fissions.

The measurement of the Pr144 2.18 MeV γ photopeak is utilized also for the direct burnup determination in irradiated rods (5). The gamma spectrum of a uranium sample irradiated about 1 year before is presented in figure 7.3. In figures 7.2 and 7.3 some other γ photopeaks are observed. However these peaks are formed by the sum of gammas of different fission products and their evaluation would require quite complicated procedures.

7.3 Gamma spectroscopy by Germanium Li-drifted detectors.

For gamma spectrometry Germanium Li-drifted detectors constructed in the laboratory (6) were used. Due to the strong activity of the samples it was preferable to employ detectors with smaller volume but better resolution. A typical detector was 1.8 cm² x 3.5 mm thick. The resolution was of the order of 3.8 KeV for the 662 KeV gamma of Cs-137.

The detector was stored and operated at liquid nitrogen temperature at vacuum from a rotary pump with an applied bias of 200 Volts. The detector was connected to a low noise charge sensitive preamplifier designed in the laboratory by R. Benoit. The preamplifier noise was of the order of 2.2 KeV at zero input capacitance.

The high resolution of Germanium Li-drifted detectors allows one to evaluate several nuclides directly from the fission product mixture. In the next part of this section the applications and results of these detectors are described.

.4 Determination of U235 and Pu239 fissions in UC specimens, irradiated by the Physical Chemistry Laboratory.

The Physical Chemistry Laboratory has irradiated two rigs containing UC specimens in the Ispra I reactor in 1964. 3 cylindrical specimens of UC containing natural Uranium -1.9 cm in diameter and 3.81 cm long - were irradiated in each rig. The 3 UC cylinders were placed at a distance of about 9 cm from each other by suitable spacers.

The rig 1 was irradiated for about 400 hours in the fuel channel n° 9 at the periphery of the Ispra I core and for three cycles of about 260 hours each in the central position of the Ispra I core. The rig 2 was irradiated in the central position of the Ispra I core for 2 cycles of about 400 hours each.

The samples to be analyzed have been removed by impact from the specimen surface, a more accurate method for the UC sampling not being available. A sketch of the rigs shows the sampling positions (Fig. 7.4). The UC samples have weights between 100 mg and 2 g.

The samples have normally been dissolved in nitric acid solution (concentration between 3 and 6 N) at a temperature of about 80°C. A few mgs of inactive carriers of Ce, Cs and Ru have been added before the dissolution.

One sample - n° 5 of RIG 1 - has been divided in two parts: the first has been dissolved in 3N HCl, the second has been dissolved in 3N HNO₃. The agreement between the activity measurements in the two solutions is within the experimental error. The experiment was performed in order to check eventual losses of Ruthenium during the nitric dissolution.

The determination of the number of fissions of U235 and Pu239 has been made by measuring the activities of the fission products Ce144, Cs137 and Ru106 by gamma spectrometry with Germanium Li-drifted detectors. A typical gamma spectrum obtained about 1 $\frac{1}{2}$ years after the end of the

irradiation and utilized for the measurement is shown in fig. 7.5. The measurement of the activities has been made by evaluating the area of the 513 KeV gamma photopeak of the Rh106 daughter of Ru106, the area of the 662 KeV gamma photopeak of the Ba137 m daughter of Cs137 and the area of the 134 KeV gamma photopeak of Ce144.

The gamma photopeaks of Rh106 and Ba137 m are shown in fig. 7.5 and the gamma photopeak of Ce144 in fig. 7.6.

The Germanium Li-drifted detector has been calibrated in efficiency by sources of known absolute activities. The absolute activity of the pair Ce144 - Pr144 has been determined by an extrapolation method applied to a $4\pi\beta-\gamma$ coincidence measurement.

A 4π gas flow counter was used for the β -ray measurement and a 3" x 3" NaI(Tl) crystal was used for the γ ray measurement.

The extrapolation method has been suggested for the Ce144 - Pr144 absolute activity measurement by H. Houtermans et al (7).

Ce144 - Pr144 sources have been prepared on VYNS films with the addition of different amounts of carrier in order to have different β efficiencies determined from the ratio between the coincidence counting N_C and the gamma counting N_γ . In fig. 7.7 the β count N_β obtained for the different sources is plotted against the β efficiency ϵ_β . The value of N_β extrapolated for $\epsilon_\beta = 1$ is the absolute activity of the pair of nuclides Ce144 - Pr144, sum of the absolute activities of the single nuclides. The measurement of the absolute activity of the standard solution has been made two times obtaining results in agreement within 1.5%.

The absolute activity measurement of the pair Ru106 - Rh106 has been determined by a similar technique. The sources have been covered by aluminium foils thick enough to stop all the betas from Ru106. Ru106 is a pure β^- emitter ($E_{Max} = 39$ KeV). Aluminium foils with different thicknesses have been utilized obtaining different β efficiencies. The N_β values have been extrapolated to $\epsilon_\beta = 1$ (fig. 7.8) obtaining the absolute activity of Rh106. Ru106 and Rh106 in equilibrium have the same activity.

Two measurements of the absolute activity of the standard solution have been made obtaining an agreement within 2%.

The absolute activity of the standard solution of Cs137 has been measured by $4\pi\beta$ counting and by γ -spectrometry with a 3" x 3" NaI(Tl) crystal with Beryllium window. In

the 4π measurement we have assumed for the β^- and conversion electron efficiency a value of 0.96. This value has been chosen on the basis of our previous experiments in beta counting.

In the γ -spectrometry measurement we have evaluated the 662 KeV gamma photopeak of the Ba137 m daughter of Cs137 utilizing the efficiency values reported by R.L. Heath (8). For the decay scheme of Cs137 - Ba137 m (fig. 7.9) we have assumed the following data (9):

95% of Cs137 decaying to Ba137 m : $e/\gamma = 0.119$

hence 10.1 conversion electrons / 100 dis

84.9 gammas / 100 dis

The agreement between the results of the two techniques is within 2%.

All the measurements with the Ge-Li drifted detectors have been made on sources of 5 ml of solution placed in small polythene bottles. The UC content in the sources was in the range of 1 - 4 mg.

The activities of Ce144, Cs137 and Ru106, corrected for decay, are reported in tables 7.1 and 7.2 for the two rigs. The decay correction of Ru106 has been calculated taking into account separately the decay of Ru106 from U235 fission and the decay of Ru106 from Pu239 fission. The fission rate of U235 can be considered to be constant during the irradiation while the fission rate of Pu239 is increasing with time. In the calculations we have utilized the following half-lives (9):

Ce144	284 days
Ce137	30 years
Ru106	365 days

Destruction by neutron capture and formation from the adjacent mass chains of the nuclides Cs137, Ce144, and Ru106 have been assumed to be negligible on the basis of the neutron cross sections reported in the literature.

The neutron capture cross section of Cs137 has been measured by D.C. Stuepegia to be 0.110 ± 0.033 barns for pile neutrons (10). The cross section for thermal neutron capture by the stable fission product Xe136, producing Xe137, father of Cs137, has been measured by M. Bresesti et al (11) to be 0.281 ± 0.028 barns. The thermal neutron capture cross section of Ce144 has been reported by P.M. Lantz to be 1.00 ± 0.1 barn (12).

The formation cross section from Ce143 (33 h half life) has been measured by L.P. Roy and L. Yaffe (13) to be 6.0 ± 0.7 barns for pile neutrons.

The thermal neutron capture cross section of Ru106 has been measured by R.D. Werner and T.A. Eastwood (14) to be 0.146 ± 0.04 barns. The formation cross section from Ru 105 has been determined by B.L. Sharma (15) to be 0.20 ± 0.02 barns for pile neutrons.

In fig. 7.10 and 7.11 the average values of the activities are plotted against the sampling positions for the two rigs.

The numbers of fissions of U235 and Pu239 have been obtained solving the systems:

$$\begin{cases} N_{Ce} = F_5 y_{Ce5} + F_9 y_{Ce9} \\ N_{Ru} = F_5 y_{Ru5} + F_9 y_{Ru9} \end{cases}$$

$$\begin{cases} N_{Cs} = F_5 y_{Cs5} + F_9 y_{Cs9} \\ N_{Ru} = F_5 y_{Ru5} + F_9 y_{Ru9} \end{cases}$$

where

N is the number of atoms formed in the fission

F is the number of fissions

y is the fission yield

5 is the index for U235

9 is the index for Pu239

In the solution of the systems the following data have been assumed for the fission yields:

$$\begin{cases} y_{Ce5} = 5.67 \% & (3) \\ y_{Cs5} = 6.05 \% & (3) \\ y_{Ru5} = 0.38 \% & (3) \end{cases}$$

$$\begin{cases} y_{Ce9} = 3.84 \% & (16) \\ y_{Cs9} = 6.43 \% & (16) \\ y_{Ru9} = 4.53 \% & (17) \end{cases}$$

In table 7.3 and 7.4 is reported the number of fissions of U235 and Pu239 for the two rigs and in fig. 7.12 and 7.13 the average number of fissions is plotted against the sampling positions for the two rigs.

The satisfactory agreement between the numbers of fissions calculated from the Ce144 and Cs137 activities allows one to conclude that the diffusion of Cs137 in this irradiation is negligible. The total error in the determination of the U235 fissions is between 5 and 10%, and in the determination of the Pu239 fissions is between 10 and 20%.

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Table 7.1: Ru-106, Cs-137, Ce-144 activities measured in samples from RIG 1.

Sample	Source	Ru-106 dis/min mg UC	Cs-137 dis/min mg UC	Ce-144 dis/min mg UC	
1	1/1	1.453 x 10 ⁷	4.152 x 10 ⁶	1.575 x 10 ⁸	
	1/2	1.528 x 10 ⁷	4.421 x 10 ⁶	1.554 x 10 ⁸	
	1/3	1.474 x 10 ⁷	4.318 x 10 ⁶	1.575 x 10 ⁸	
Average value		1.485 x 10 ⁷	4.297 x 10 ⁶	1.568 x 10 ⁸	
2	2/1	1.466 x 10 ⁷	4.213 x 10 ⁶	1.455 x 10 ⁸	
	2/2	1.475 x 10 ⁷	4.160 x 10 ⁶	1.463 x 10 ⁸	
	2/3	1.462 x 10 ⁷	4.255 x 10 ⁶	1.476 x 10 ⁸	
Average value		1.468 x 10 ⁷	4.209 x 10 ⁶	1.465 x 10 ⁸	
5	HCl { 1	1.736 x 10 ⁷	4.656 x 10 ⁶	1.655 x 10 ⁸	
		2	1.778 x 10 ⁷	4.571 x 10 ⁶	1.606 x 10 ⁸
	HNO ₃ { 5	1.811 x 10 ⁷	4.491 x 10 ⁶	1.601 x 10 ⁸	
		6	1.819 x 10 ⁷	4.447 x 10 ⁶	1.630 xx 10 ⁸
		Average value		1.786 x 10 ⁷	4.541 x 10 ⁶
6	3	2.037 x 10 ⁷	5.035 x 10 ⁶	1.716 x 10 ⁸	
	4	1.956 x 10 ⁷	4.916 x 10 ⁶	1.667 x 10 ⁸	
Average value		1.992 x 10 ⁷	4.975 x 10 ⁶	1.691 x 10 ⁸	
7	7/1	1.953 x 10 ⁷	5.114 x 10 ⁶	1.807 x 10 ⁸	
	7/2	1.898 x 10 ⁷	4.685 x 10 ⁶	1.692 x 10 ⁸	
	7/3	1.962 x 10 ⁷	4.876 x 10 ⁶	1.664 x 10 ⁸	
Average value		1.938 x 10 ⁷	4.892 x 10 ⁶	1.721 x 10 ⁸	
9	9/1	1.887 x 10 ⁷	4.729 x 10 ⁶	1.738 x 10 ⁸	
	9/2	1.866 x 10 ⁷	4.586 x 10 ⁶	1.762 x 10 ⁸	
	9/3	1.961 x 10 ⁷	4.838 x 10 ⁶	1.721 x 10 ⁸	
Average value		1.905 x 10 ⁷	4.718 x 10 ⁶	1.740 x 10 ⁸	

Table 7.2: Ru-106, Cs-137, Ce-144 activities measured in samples from RIG - 2.

Sample	Source	Ru-106 dis/min mg UC	Cs-137 dis/min mg UC	Ce-144 dis/min mg UC
A ₂ B ₃	A/1	1.188 x 10 ⁷	3.491 x 10 ⁶	1.241 x 10 ⁸
	A/2	1.182 x 10 ⁷	3.562 x 10 ⁶	1.257 x 10 ⁸
	A/3	1.215 x 10 ⁷	3.617 x 10 ⁶	1.238 x 10 ⁸
Average value		1.195 x 10 ⁷	3.557 x 10 ⁶	1.245 x 10 ⁸
B ₂ H ₄	B/1	1.597 x 10 ⁷	3.867 x 10 ⁶	1.378 x 10 ⁸
	B/2	1.548 x 10 ⁷	3.846 x 10 ⁶	1.372 x 10 ⁸
	B/3	1.546 x 10 ⁷	3.897 x 10 ⁶	1.334 x 10 ⁸
Average value		1.564 x 10 ⁷	3.870 x 10 ⁶	1.361 x 10 ⁸
B ₂ M ₅	M/1	1.373 x 10 ⁷	3.854 x 10 ⁶	1.375 x 10 ⁸
	M/2	1.468 x 10 ⁷	3.849 x 10 ⁶	1.384 x 10 ⁸
	M/3	1.382 x 10 ⁷	3.834 x 10 ⁶	1.393 x 10 ⁸
Average value		1.408 x 10 ⁷	3.846 x 10 ⁶	1.384 x 10 ⁸
B ₂ B ₆	1*	1.572 x 10 ⁷	4.066 x 10 ⁶	1.438 x 10 ⁸
	2*	1.481 x 10 ⁷	4.004 x 10 ⁶	1.411 x 10 ⁸
Average value		1.526 x 10 ⁷	4.035 x 10 ⁶	1.424 x 10 ⁸

Table 7.3 : U-235 and Pu-239 Fissions measured in samples from RIG 1.

Sample	U-235 Fissions / mg UC			Pu-239 Fissions / mg UC		
	pair { Ce-144 Ru-106	pair { Cs-137 Ru-106	average value	pair { Ce-144 Ru-106	pair { Cs-137 Ru-106	average value
1	1.559×10^{15}	1.483×10^{15}	<u>1.521×10^{15}</u>	1.180×10^{14}	1.245×10^{14}	<u>1.212×10^{14}</u>
2	1.447×10^{15}	1.451×10^{15}	<u>1.449×10^{15}</u>	1.245×10^{14}	1.242×10^{14}	<u>1.243×10^{14}</u>
5	1.581×10^{15}	1.518×10^{15}	<u>1.549×10^{15}</u>	1.665×10^{14}	1.713×10^{14}	<u>1.689×10^{14}</u>
6	1.635×10^{15}	1.665×10^{15}	<u>1.650×10^{15}</u>	1.964×10^{14}	1.939×10^{14}	<u>1.951×10^{14}</u>
7	1.675×10^{15}	1.748×10^{15}	<u>1.711×10^{15}</u>	1.840×10^{14}	1.869×10^{14}	<u>1.854×10^{14}</u>
9	1.699×10^{15}	1.573×10^{15}	<u>1.636×10^{15}</u>	1.765×10^{14}	1.871×10^{14}	<u>1.818×10^{14}</u>

Table 7.4: U-235 and Pu-239 Fissions Measured in samples from RIG 2.

Sample	U-235 Fissions / mg UC			Pu-239 Fissions / mg UC		
	pair { Ce-144 Ru-106	pair { Cs-137 Ru-106	average value	pair { Ce-144 Ru-106	pair { Cs-137 Ru-106	average value
A ₂ B ₃	1.236 x 10 ¹⁵	1.235 x 10 ¹⁵	<u>1.235 x 10¹⁵</u>	9.647 x 10 ¹³	9.654 x 10 ¹³	<u>9.650 x 10¹³</u>
B ₂ H ₄	1.321 x 10 ¹⁵	1.455 x 10 ¹⁵	<u>1.388 x 10¹⁵</u>	1.513 x 10 ¹⁴	1.400 x 10 ¹⁴	<u>1.456 x 10¹⁴</u>
B ₂ M ₅	1.365 x 10 ¹⁵	1.448 x 10 ¹⁵	<u>1.406 x 10¹⁵</u>	1.212 x 10 ¹⁴	1.365 x 10 ¹⁴	<u>1.288 x 10¹⁴</u>
B ₂ B ₆	1.395 x 10 ¹⁵	1.518 x 10 ¹⁵	<u>1.456 x 10¹⁵</u>	1.386 x 10 ¹⁴	1.283 x 10 ¹⁴	<u>1.334 x 10¹⁴</u>

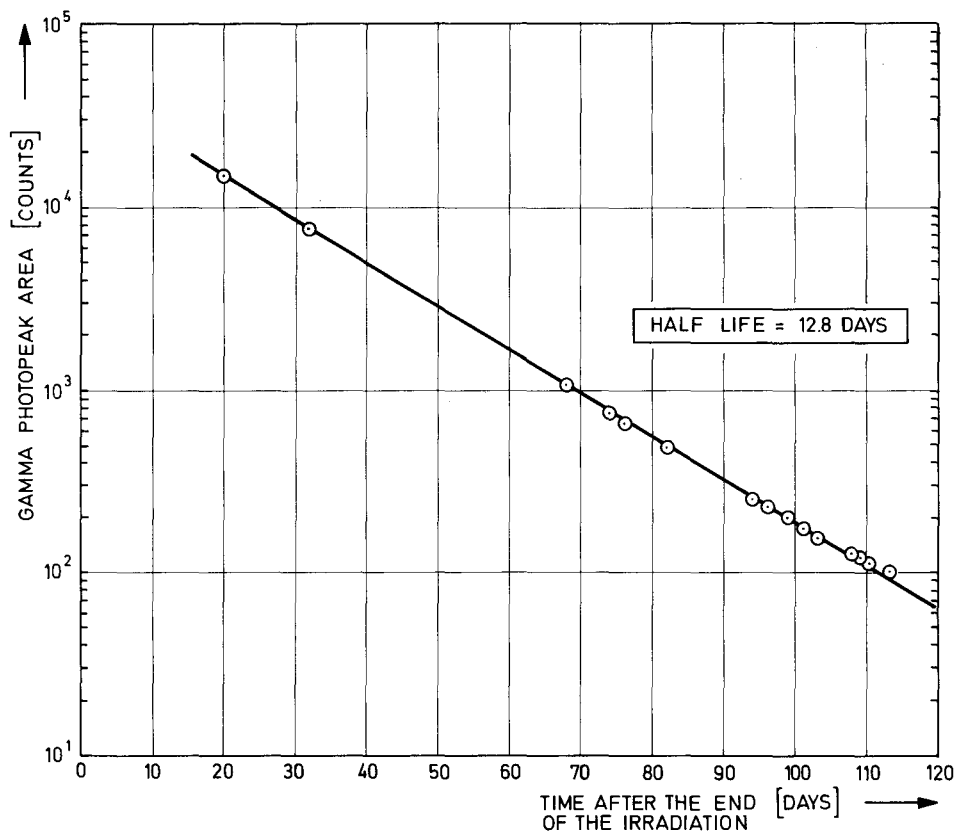


Fig. 7.1 DECAy OF THE 1.60 MeV La-140 GAMMA PHOTOPEAK EVALUATED IN A FISSION PRODUCT MIXTURE BY A 3"x3" NaI (TI) CRYSTAL

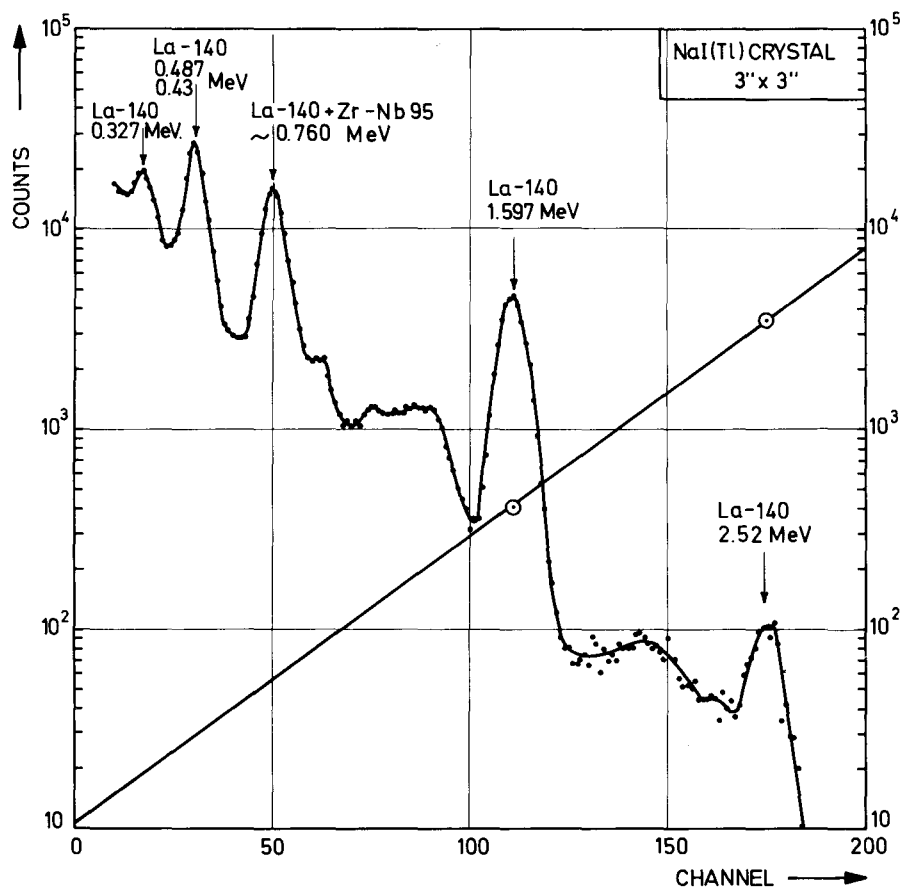


Fig. 7.2 GAMMA SPECTRUM FOR A FISSION PRODUCT MIXTURE ABOUT 1 MONTH AFTER THE END OF THE IRRADIATION

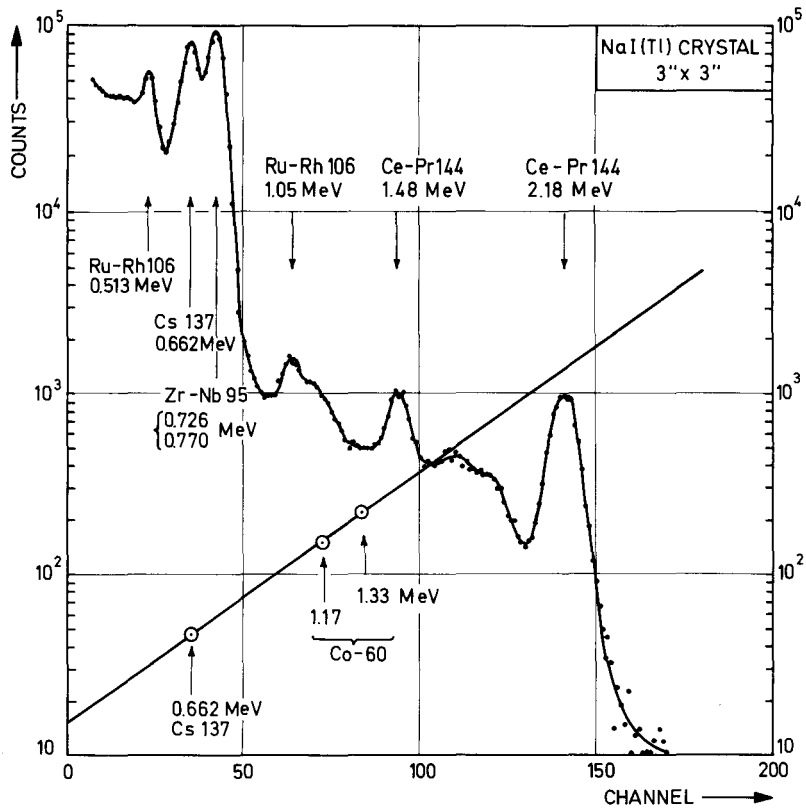


Fig. 7.3 GAMMA SPECTRUM FOR A FISSION PRODUCT MIXTURE ABOUT 1.5 YEARS AFTER THE END OF THE IRRADIATION

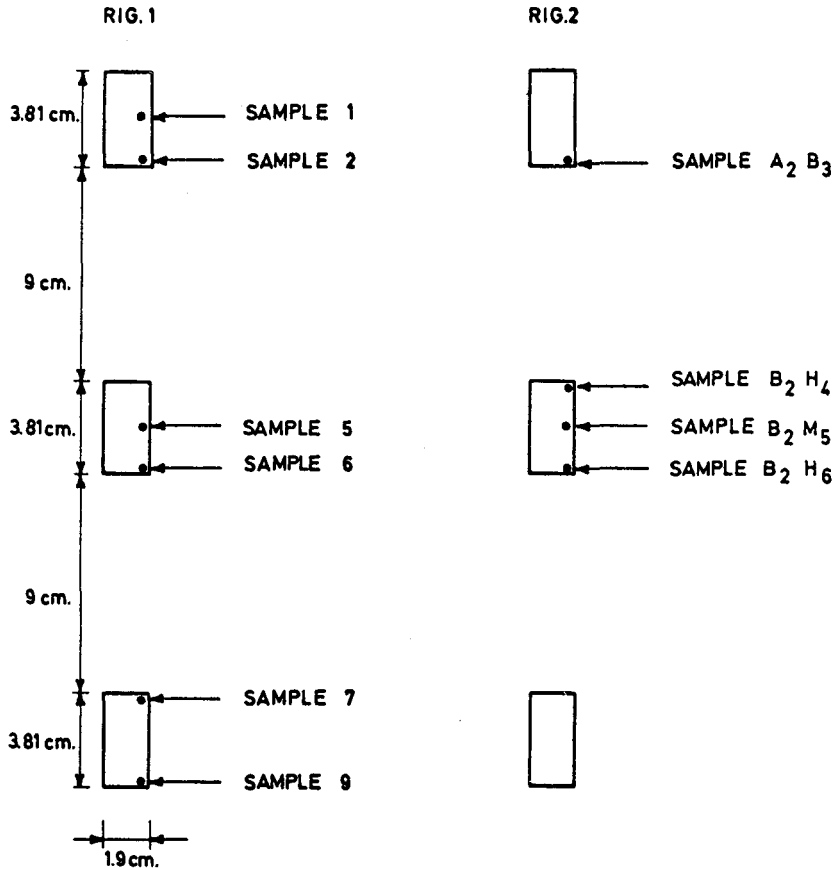


Fig. 7.4 SKETCH OF THE RIGS WITH THE SAMPLING POSITION

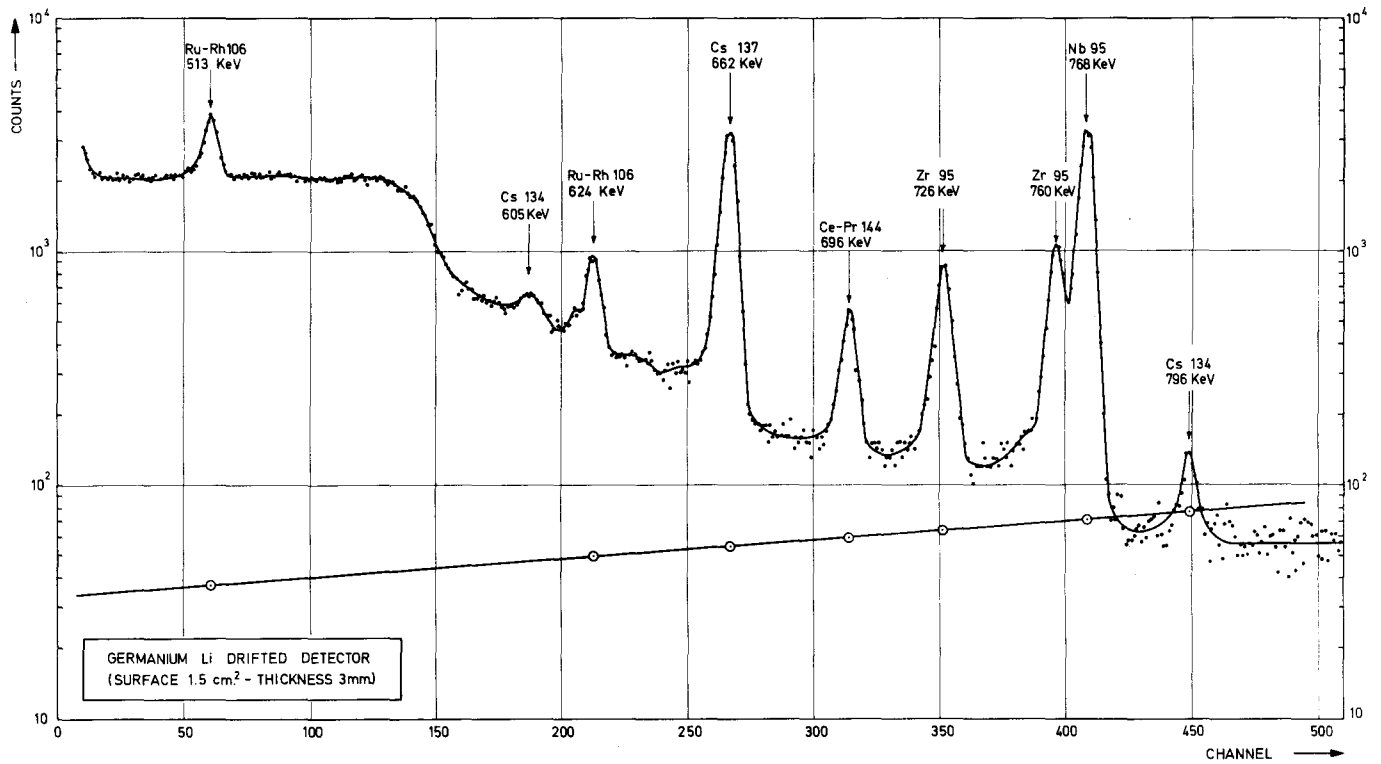


Fig. 7.5 GAMMA SPECTRUM OF A FISSION PRODUCT MIXTURE ABOUT 1.5 YEARS AFTER THE END OF THE IRRADIATION

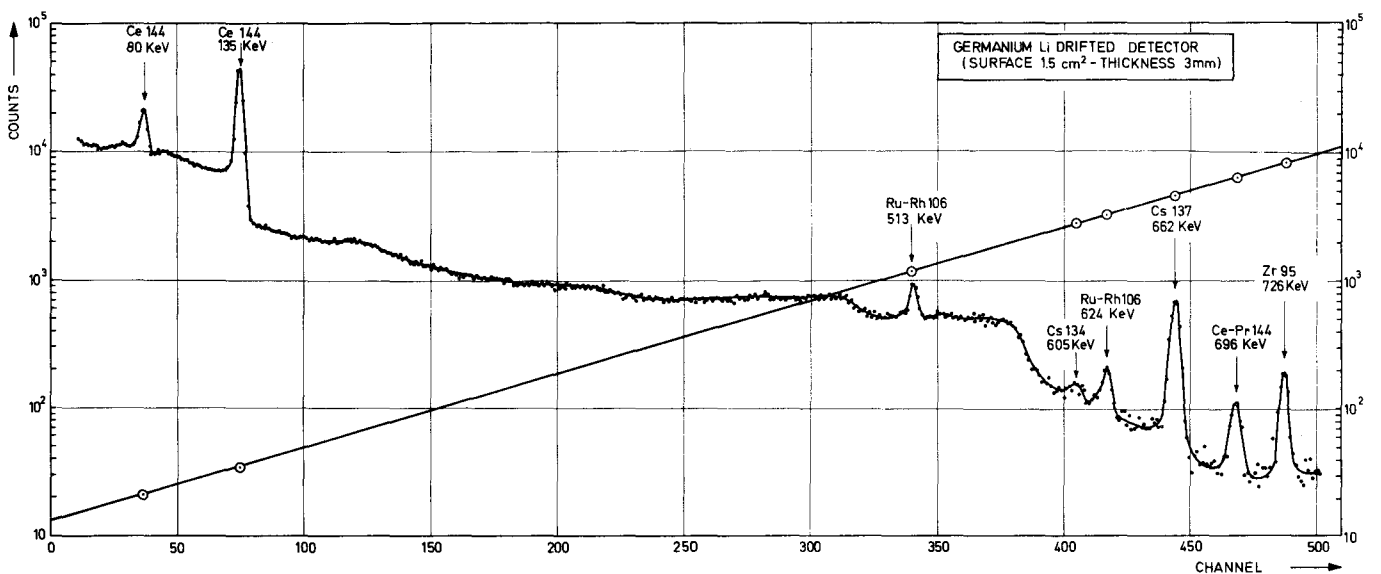


Fig. 7.6 GAMMA SPECTRUM FOR A FISSION PRODUCT MIXTURE ABOUT 1.5 YEARS AFTER THE END OF THE IRRADIATION

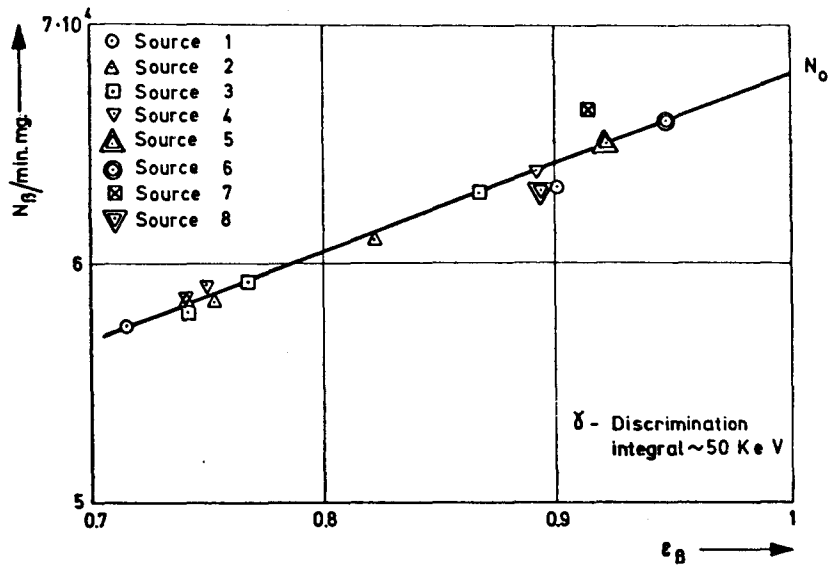


Fig. 7.7- SPECIFIC β -COUNTING RATE AS A FUNCTION OF THE β -EFFICIENCY FOR DIFFERENT SOURCES OF Ce - Pr 144

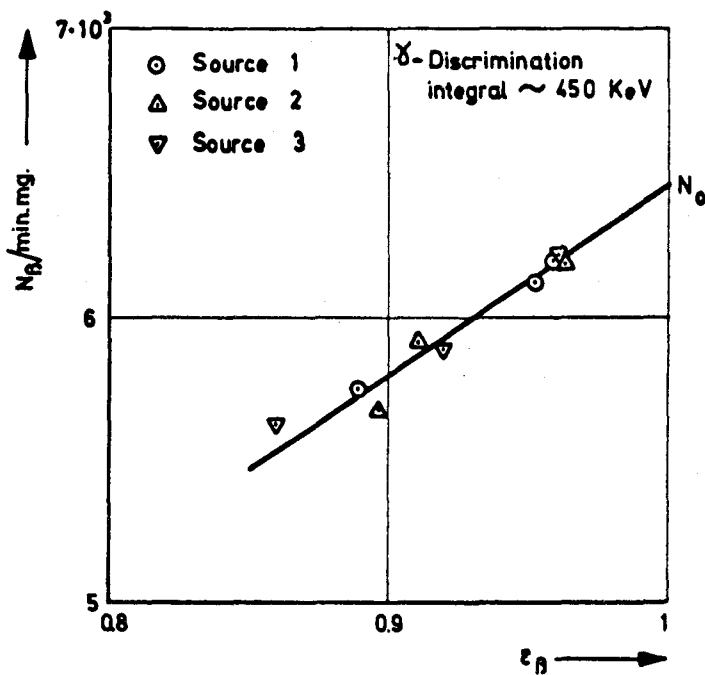


Fig. 7.8- SPECIFIC β -COUNTING RATE AS A FUNCTION OF THE β -EFFICIENCY FOR DIFFERENT SOURCES OF Ru-Rh 106

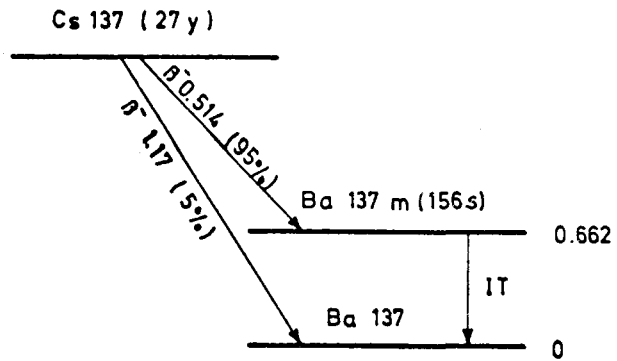


Fig. 7.9- DECAY SCHEME OF Cs 137 - Ba 137m.

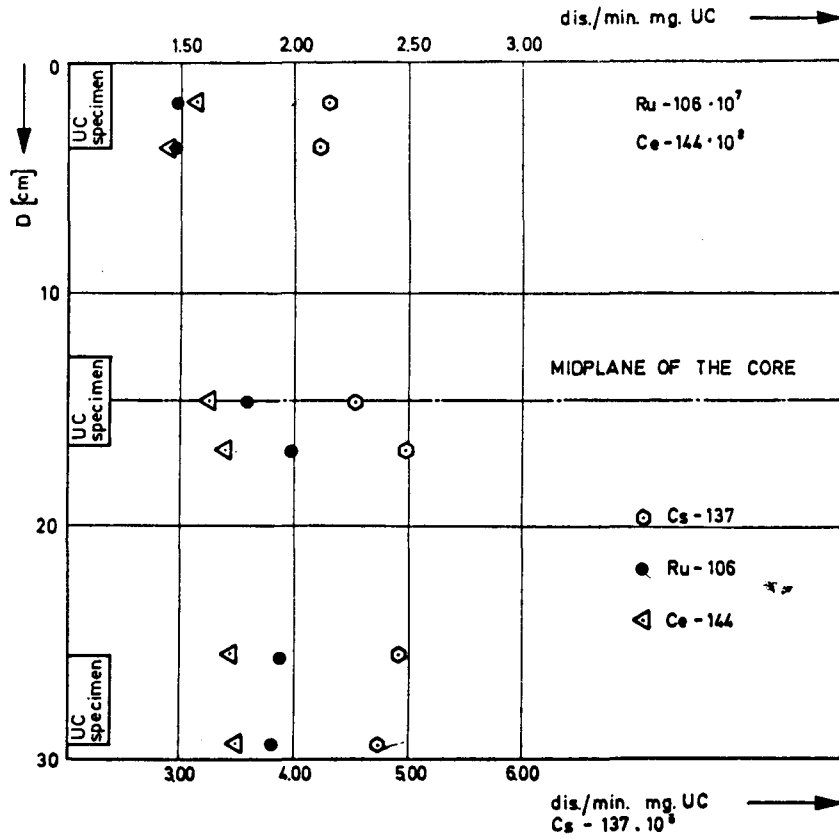


Fig. 7.10- Ru-106, Cs-137, Ce-144 ACTIVITIES MEASURED IN SAMPLES FROM FIG.1

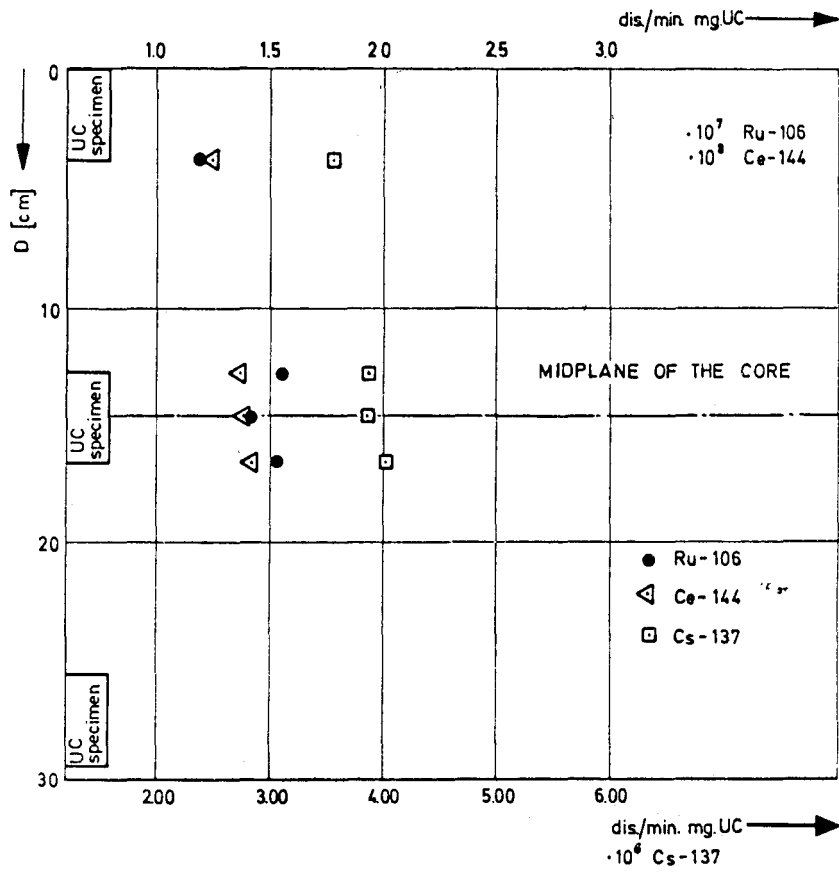


Fig. 7.11 - Ru-106, Cs-137, Ce-144 ACTIVITIES
MEASURED IN SAMPLES FROM RIG 2

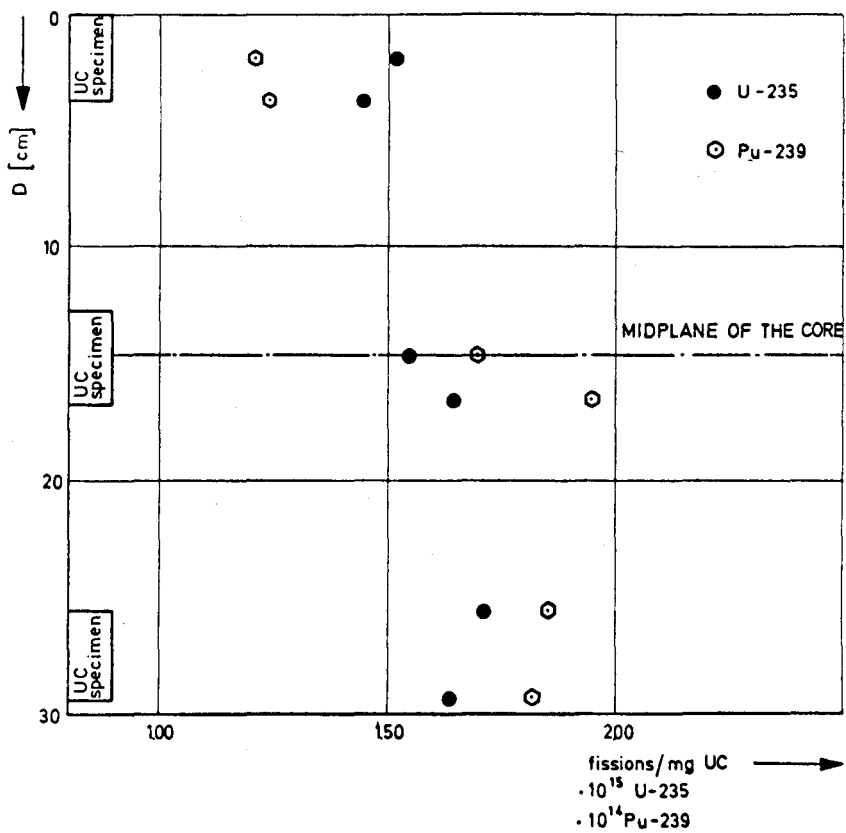


Fig. 7.12 - U-235 AND Pu-239 FISSIONS MEASURED IN
SAMPLES FROM RIG.1

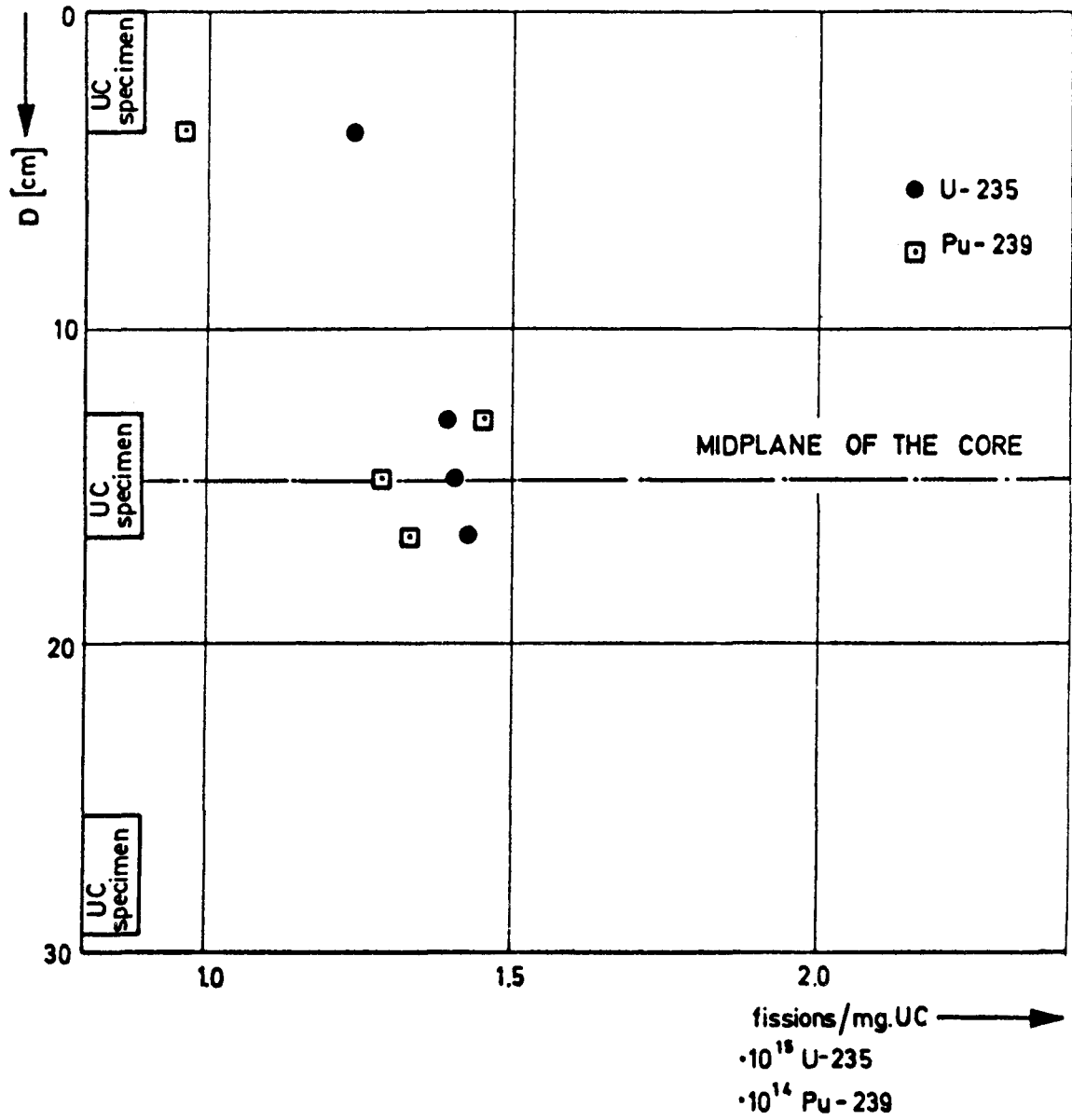


Fig. 7.13 - U-235 AND Pu-239 FISSIONS MEASURED IN SAMPLES FROM FIG. 2

8 - CHEMICAL PROCEDURE IN THE DETERMINATION OF THE STABLE FISSION PRODUCT Nd 148.

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8.1 Introduction.

The determination of a stable fission product by mass spectrometry using the isotope dilution technique allows one to operate with a much higher accuracy than the measurement of a radioactive fission product.

Nd148 is the most interesting stable fission product and its measurement has been studied by B.F. Rider et al (1) (2). Nd148 has a low tendency to migrate, has a low destruction cross section and a low formation cross section from adjacent mass chains, and has a similar fission yield in the fission of U235 and Pu239. Moreover, the Neodymium element has a nuclide - Nd142 - not formed in fission which permits correction for natural contamination. Enriched Nd150 was selected by B.F. Rider et al (1) as the diluting isotope.

In the Nuclear Chemistry Laboratory a radiochemical procedure has been set up for the separation of individual rare earths produced in fission. This separation was utilized in the determination of fission yields in the fission of Th232 by fast neutrons.

The same procedure could be utilized for the separation of the Neodymium isotopes from other fission products if the determination of Nd148 appears to be convenient.

8.2 Experimental.

The rare earths as a group have been purified from other fission products by a series of radiochemical operations (3). These include a zirconium phosphate scavenging precipitation, a precipitation of rare-earths fluorides, a barium sulphate scavenging precipitation, precipitation of rare-earth hydroxides, and passage of the rare earths through a small anion-exchange column in concentrated hydrochloric acid.

The separation of individual rare earths has been made utilizing the method proposed by K. Wolfsberg (4). This method consists of elution through cation-exchange resin columns operated at room temperature with α -hydroxy-isobutyrate solutions at pH increasing from 3.4 to 4.2. An elution curve

obtained in the separation of the rare earths produced in the Th²³² fission is shown in fig. 8.1.

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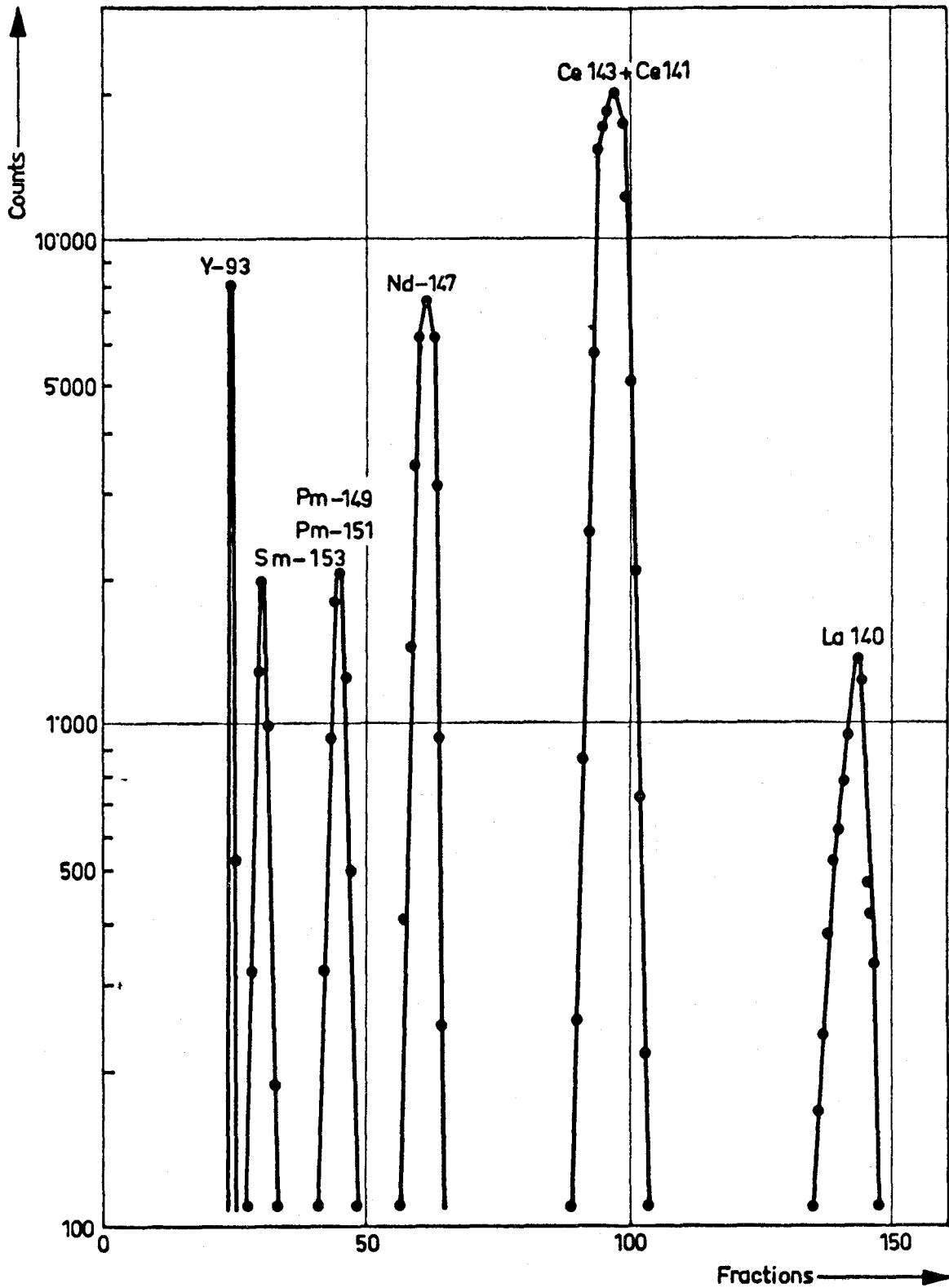


Fig 8.1 - SEPARATION OF THE RARE EARTHS BY CATION EXCHANGE
WITH α -HYDROXY ISOBUTYRIC ACID

9 - DETERMINATION OF Pu239 + Pu240 ACTIVITY BY α SPECTROMETRY

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9.1 Introduction.

From the measurement of the Pu239 + Pu240 activity it is possible to obtain the burnup value only when accurate information on the neutron spectrum is available.

In Section 1 calculations of neutron spectra are reported for a UC specimen to be irradiated in the DIRCE loop of the Ispra I reactor. From the neutron spectrum the parameter β , defined as the ratio of total to thermal + $1/v$ neutron captures in U238, is calculated. The utilization of this parameter in the burnup calculation from the Pu239 + Pu240 activity is reported in Reference (1).

The alphas from Pu239 and Pu240 have very similar energies and cannot be distinguished by α -spectrometry with ionization chambers or semiconductor detectors available in the Nuclear Chemistry Laboratory.

Preliminary experiments for the determination of the Pu239 + Pu240 activity have been made utilizing a method based on isotopic dilution with Pu238 and α -spectrometry with semiconductor detectors. The utilization of this method is more complicated if Pu238 is present in the fuel to be analyzed. The energy and the intensity of the main alphas of Pu238, Pu239 and Pu240 are reported in Table 9.1.

Table 9.1.: Energy and intensity of the alphas of Pu238, Pu239, Pu240.

Pu238 alphas		Pu239 alphas		Pu240 alphas	
Energy (MeV)	Intensity %	Energy (MeV)	Intensity %	Energy (MeV)	Intensity %
5.491	71.1	5.147	72.5	5.159	75.5
5.448	28.7	5.134	16.8	5.115	24.4
5.352	0.13	5.096	10.7	5.014	0.091
5.208	0.005	5.064	0.037	4.952	0.0027
		4.999	0.013	4.924	0.0031
		4.917	0.005	4.851	0.0032

The method requires, after the addition of Pu238, a purification of the Plutonium mixtures from Uranium, Americium and fission products. The source for the α -spectrometry is prepared by electrodeposition. An alpha spectrum obtained by

semiconductor detectors of the mixture of Pu238 + Pu239 + Pu240, separated from irradiated uranium carbide, is shown in fig. 9.1. The study of the details of the experimental procedures of purification and electrodeposition of plutonium has still to be completed.

In the experiments performed plutonium amounts of a few μg were separated from uranium carbide amounts of a few mg. The uranium carbide has been irradiated for about 30 days in a thermal flux of $\sim 5 \times 10^{13}$ n/cm² sec.

9.2 Isotopic dilution with Pu238.

Pu238 seems to be a very convenient isotope for the isotopic dilution of the Pu239 + Pu240 pair. This isotope can be purchased at a low price with high radiochemical purity. The α -ray energies of Pu238 are quite different from the α -ray energies of Pu239 and Pu240 and this facilitates the α spectrometry.

The radiochemical purity of Pu238, relative to the alpha emission, has been investigated by alpha and gamma spectrometry. In the alpha spectrum only α -rays with energies similar to the Pu238 energies have been observed. However the alphas of Am241 cannot be distinguished from the alphas of Pu238. The presence of Am241 has been excluded by γ spectrometry measurements by Germanium Li-drifted detectors.

In the isotopic dilution a known absolute activity of Pu238 has to be added to the solution. The standardization of an alpha decaying isotope is normally performed by a 4π counter. Experiments for the evaluation of the error introduced in this measurement are in progress.

9.3 Chemical purification of Plutonium.

The plutonium mixture has to be purified from the fission products in order to reduce the β and γ activity of the source, and from the Uranium in order to obtain a source with low deposit as required for the α spectrometry. In the method utilized the higher oxidation states of plutonium are reduced to Pu (III) followed by conversion to Pu(IV), which is extractable with thenoyltrifluoroacetone (TTA). The TTA phase is stripped of the plutonium with hydrochloric acid.

Reagents.

Hydroxylamine Hydrochloride Solution, 5M. Dissolve 347.5 g of $\text{NH}_2\text{OH} \cdot \text{HCl}$ in 1 liter of water.

Ferric Nitrate Solution, 0.1 M. Dissolve 35 g of $\text{Fe}(\text{NO}_3)_3 \cdot 6 \text{H}_2\text{O}$ in 1 liter of water.

Sodium Nitrite Solution, 3M. Dissolve 20.7 g of NaNO_2 in

100 ml of water. This solution is not stable and must be prepared fresh daily before use.

Thenoyltrifluoroacetone (TTA) Solution, 0.5 M. Dissolve 5.555 g of TTA in 50 ml of xylene.

Procedure.

Make the solution of irradiated uranium about 1M with HNO_3 . Transfer about 1 ml of the solution in an extraction vial, add 2 drops of 0.1 M $\text{Fe}(\text{NO}_3)_3 \cdot 6 \text{H}_2\text{O}$ solution and 2 drops of 5M $\text{NH}_2\text{OH} \cdot \text{HCl}$ solution and stir by hand for 5 min.

Add 10 drops of 3M NaNO_2 solution and stir by hand for 5 min. Add by means of a pipet 1 - 2 ml of 0.5 M TTA in order to have a ratio between the volumes of organic phase and aqueous phase of about 1 and stir by hand for 15 min.

Centrifuge for 5 min and separate the organic phase. Wash the organic phase twice with 1ml of 1M HNO_3 and strip the plutonium with 10 M HCl .

9.4 Electrodeposition of Plutonium.

The electrodeposition of Plutonium has been made following the procedure of R.F. Mitchell (2).

The electrodeposition cell consists of a metal frame at the bottom of which a tantalium disk is placed to serve as the cathode. A neoprene gasket and a plexiglas cylinder are secured on top of the disk. A rotating platinum electrode is the anode. The electrodeposition was made from solutions of about 5 ml volume and with a distance between the electrodes of about 5 mm. A current density of 0.5 Amp/cm^2 was utilized.

9.5 Alpha spectrometry using semiconductor detectors.

Alpha spectrometry was performed using Silicon surface barrier detectors constructed in the laboratory. Due to the low activity of the sources it was necessary to use large area detectors to reduce the counting time.

Detectors were prepared from $6000 \Omega\text{-cm}$ Silicon and they were of the araldite encapsulated type; the useful area was 2.5 cm^2 . The resolution was of the order of 35 KeV at room temperature.

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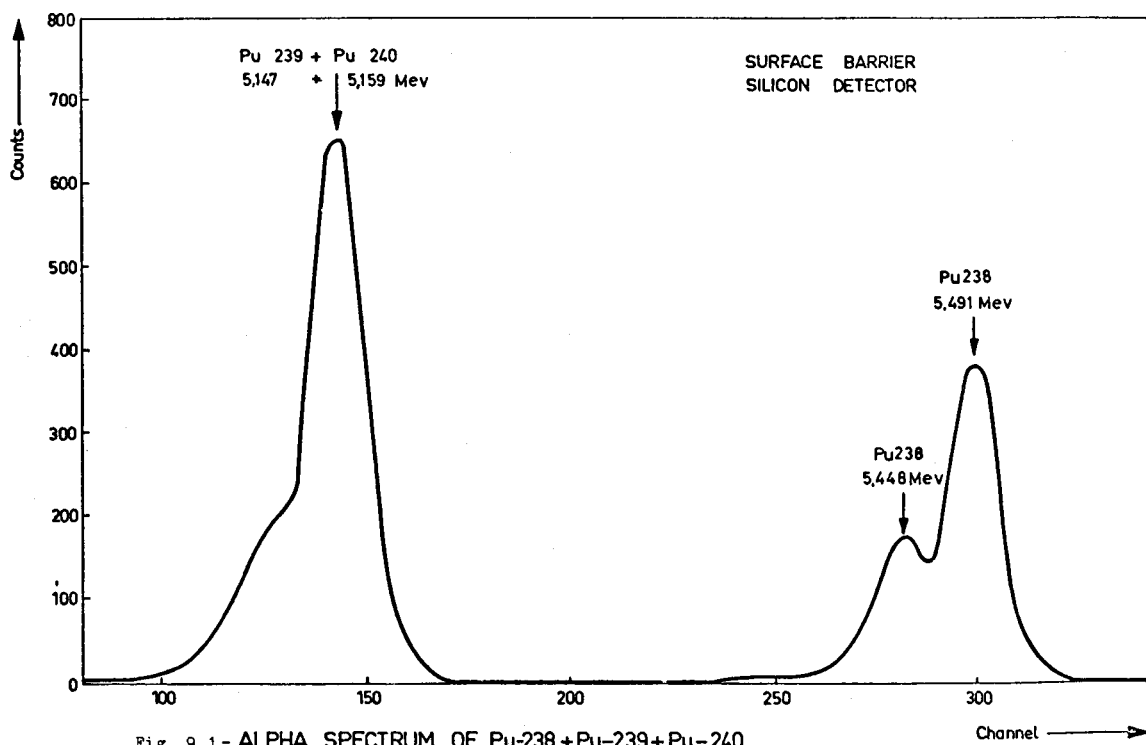


Fig. 9.1 - ALPHA SPECTRUM OF Pu-238 + Pu-239 + Pu-240

10 - NEUTRON FLUX MEASUREMENTS AT THE SURFACE OF UC SPECIMENS.

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10.1 Introduction.

The integrated flux received by an irradiated specimen can be determined by flux monitoring the surface of the specimen when the ratio between the surface flux and the average flux in the volume is known.

Preliminary calculations have been performed (see section 1) for the DIRCE specimens (UC cylindrical samples, containing natural uranium, 15 cm high by 2.5 cm in diameter) to be irradiated in the central position of the Ispra 1 core. These calculations have shown that the flux depression in the radial direction is relatively small. For the thermal flux the ratio of average to superficial flux is about 0.9 and is even higher for the higher neutron energies. This means that the corrections to be taken into account for flux depression are of the order of 10%.

When the method, based on surface flux measurements and related calculations, is proven this kind of determination will permit one to eliminate the troublesome operations of sampling the irradiated materials and eliminate the radio-chemical procedures required by the other methods. Furthermore, by a suitable distribution of monitors it is possible to obtain information about the flux received by all the parts of the specimen. This information can be obtained in a much shorter time than the information from other measurements.

The flux monitoring seems to be particularly useful for the DIRCE experiment where several short period irradiations are planned. By the analysis of the flux monitor data it will be possible to know quickly about eventual modifications in the irradiation of the different parts of the specimen.

In a more general way the flux determination at the surface of the specimen will give an indication of the most suitable way of sampling the fuel for other kinds of burnup determinations. The flux monitoring method would yield great errors if the neutron absorption by fission products were important. This absorption should not be important for an irradiation of less than a few thousand MWd/t. For instance in the comparison of methods of determining burnup performed

by R.G. Hart et al. (1), the burnup values obtained with cobalt monitoring are in good agreement with the value obtained by other methods for irradiations of 3000 MWd/t.

10.2 General Information about the flux monitors.

The measurement of neutron fluxes will be performed using monitors made of a dilute alloy of Cobalt and Aluminium. Cobalt is utilized as a dilute alloy with Aluminium in order to eliminate self-shielding effects for thermal and epithermal neutrons, requiring corrections which can not always be evaluated with good precision. The low content of Cobalt in the alloy also reduces the Co60 activity that can be quite troublesome to handle in long irradiations of pure Cobalt.

The activity of a Cobalt monitor at time T, after an irradiation time t, is related to integrated flux ω as shown in equation [1].

$$n v_0 t = \omega \times 10^{21} = \frac{A M D}{W N \hat{\sigma} \lambda} \quad [1]$$

where

- A/W is the specific activity of the cobalt monitor
- N is Avogadro's number
- M is the atomic weight of cobalt
- $n v_0 t$ is the integrated 2200 m/s flux = $\omega \times 10^{21}$
- $\hat{\sigma}$ is the effective cross section of Co59 for use with the 2200 m/s flux
- λ is the disintegration constant of Co60 = $4.168 \times 10^{-9} \text{sec.}^{-1}$ corresponding to a half-life of 5.27 years.
- D is a correction factor for the Co60 decay and burnup by neutron capture. This second process is important only for high values of ω

C.H. Westcott et al. (2) have shown that $\hat{\sigma}$ can be conveniently represented by equation [2].

$$\hat{\sigma} = \sigma_0 (g + r \sqrt{T/T_0} s_0) \quad [2]$$

where

- σ_0 is the 2200 m/s neutron capture cross section of Co59 (= 38.0 barns)
- g is a factor to allow for the departure of the cross section from $1/v$ in the thermal region of the neutron spectrum (= 1 for Co59)
- $r \sqrt{\frac{T}{T_0}} s_0$ is a factor to allow for the epithermal component of the neutron spectrum and the non $\frac{1}{v}$ behaviour of the cross section.

The quantity s_0 for Co59 has been assumed equal to 1.58 .
The factor $r \sqrt{\frac{T}{T_0}}$ is usually obtained from Cd-ratio mea-

surements. Unfortunately, Cd-ratio measurements are impractical at the surface of a fuel specimen.

Preliminary calculations of $r \sqrt{\frac{T}{T_0}}$ have been made (see Section 1) for the DIRCE irradiations.

R.G. Hart et al. (3) have suggested the use of pairs of cobalt and silver monitors for the measurement of $r \sqrt{\frac{T}{T_0}}$ for

long irradiations. Silver has been chosen because the neutron capture reaction in Ag109 leads to an isomeric state of Ag110 with a 253 day half life, which allows one to perform irradiations of a few months without large decay corrections. Ag109 also has a strong absorption resonance at 5.19 eV which makes the epithermal activation important in the total activation. Furthermore, the reactor neutron capture cross section of the 253 day Ag110 m has been measured by R.G. Hart and R.L. Graham (4) and is equal to 82 barns: thus the correction for the neutron capture will be small, even for long irradiations.

Experimentally the method suggested by R.G. Hart et al. (3) consists of a measurement, for both Co59 and Ag109, of the ratio between the activation rate in the unknown spectrum and the activation rate in a spectrum where $r \sqrt{\frac{T}{T_0}}$ is known

from cadmium ratio determinations. Only relative measurements are required. A detailed description of the method can be found in reference (3).

However silver, as a monitor for the resonance region, has one drawback. The 5.19 eV resonance of Ag109 is situated near that of U238 at 6.7 eV; this may cause an important flux depression in the region of 5.19 eV (5). A theoretical calculation of the correction, taking into account the geometrical arrangement (dimensions and distance of the absorber - U238 - and monitor - Ag109), seems quite difficult.

The value of $r \sqrt{\frac{T}{T_0}}$ obtained by the activation of cobalt-silver pairs on the surface of UC contains some uncertainty. However, high precision is not required in these measurements. In any event, the use of the cobalt-silver pairs will be useful in testing the nuclear codes utilized in the calculation of the neutron spectra (see section 1) which we will have in preliminary irradiations to be performed in DIRCE using samples containing lead in place of UC. Under these conditions self-shielding effects in the region of the Ag109 resonance do not interfere and a direct comparison is possible with the results obtained by nuclear codes.

In order to avoid the uncertainties present when using silver monitors, we have tried to find another isotope more suitable than Ag109 for the resonance flux measurement. It is necessary to find an isotope which upon neutron capture yields a radioactive nuclide with a rather long half life, is easy to measure, and has a small neutron capture cross-section. Furthermore, this isotope must have rather important resonances in the epithermal region in order that the epithermal activation not be negligible in comparison with the total activation. These resonances must not be too near in energy to the U238 resonances. Keeping these considerations in mind, we found that the thulium isotope, Tm 169, being the only naturally occurring isotope of this element, corresponds to all the requirements stated above. The half life of Tm 170, which is formed upon neutron capture, is 127 days. Its thermal neutron capture cross section is 150 barns (6). These nuclear data are less favorable than the nuclear data of Ag109 because the half-life is shorter and the thermal neutron cross section is higher. Therefore the use of this monitor will be suitable for shorter irradiations. On the other hand the resonance absorption energies are much better separated from those of U238 than in the case of silver. These are shown in the following table:

Table 10.1: Capture resonance energies of U238 and Tm169 and resonance integral for Tm169.

E_r U238(eV)	E_r Tm169(eV)	Contribution to the resonance integral of Tm169 (barns)
6.68	3.92	1363
10.2	14.4	52.3
21.0	17.6	20.1
36.8	29.1	0.89
66.3	35.2	19.2

The values reported in the last column of the table have been calculated according to the following equation [3]:

$$RI = \frac{4090 g}{\Gamma} \frac{\Gamma_\gamma \Gamma_n}{E_r^2} \quad [3]$$

where g has been taken equal to $\frac{1}{2}$

Γ is the total resonance width in mV
 Γ_γ is the partial resonance width for neutron capture in mV

Γ_n is the partial resonance width for scattering in mV
 E_r is the resonance energy in eV

Tm169 has many other resonances which are not listed in Table 10.1 whose contributions to the total resonance integral are, however, negligible. As can be seen in table 10.1 the greatest part of the resonance integral is due to the resonance at 3.92 eV, which is fairly well separated from the 6.68 eV resonance of U238.

Preliminary experiments will be done on the use of the neutron capture reaction in Tm169. If these experiments give favorable results, an alloy of aluminium with low Thulium content will be prepared. The half life of Tm170 - 127 days - is sufficiently long for monitoring the DIRCE irradiations which last for a duration of a few weeks.

10.3 Preparation and analysis of Co/Al and Ag/Al alloys.

The "Istituto Sperimentale per i Metalli Leggeri" in Novara has prepared two Co/Al alloys with a Cobalt content of about 1% and 0.1%, and one Ag/Al alloy with a Silver content of about 0.1%. The alloys have been prepared by utilizing high purity aluminium, cobalt and silver.

In order to check the uniformity of the alloy composition, and to determine the cobalt and silver content, analyses have been performed in the Analytical Group of the "Istituto Sperimentale per i Metalli Leggeri" and in the Analytical Chemistry Laboratory and the Nuclear Chemistry Laboratory of CCR Euratom Ispra. In all the laboratories the composition of the alloys has been checked to be uniform within the experimental uncertainty of the methods.

The values determined for the cobalt and silver content in the alloys are reported in Tables 10.2 and 10.3. The errors reported are at the 95% confidence level.

Table 10.2: Results of the analysis of the Co/Al alloys.

Laboratory	Method	Cobalt content in %	
		alloy 1%	alloy 0.1%
Analytical Group of ISML NOVARA	Spectrophotometry	0.975(*)	0.1004 (*)
Analytical Chemistry CCR EURATOM	Spectrophotometry		
	Method 1	0.932 ± 0.005	0.0978 ± 0.0006
	Method 2	0.935 ± 0.004	0.0976 ± 0.0014
Nuclear Chemistry CCR EURATOM	Neutron Activation	0.960 ± 0.013	0.100 ± 0.002

* Average of 2 measurements in agreement within 1%.

Table 10.3 : Results of the analysis of the Ag/Al alloy.

Laboratory	Method	Silver content in %
Analytical Group of ISML NOVARA	Gravimetry	0.0985 \pm 0.0026
Analytical Chemistry CCR EURATOM	Spectrophotometry	0.0997 \pm 0.0003
Nuclear Chemistry CCR EURATOM	Neutron Activation	0.1017 \pm 0.0017

The results obtained in the analysis of the Co/Al alloys are in satisfactory agreement. The cobalt content will be assumed to be 0.95 \pm 0.015 % and 0.099 \pm 0.001% in the two alloys respectively.

The results obtained in the analysis of the Ag/Al alloy are in good agreement. The silver content in the alloy will be assumed to be 0.100 \pm 0.001%, the average value of the determinations in different laboratories. This value is very similar to the value obtained in the most careful determination 0.0997 \pm 0.0003%.

We wish to point out that the knowledge of the composition of the Co/Al alloys is important because the error in the cobalt content is directly transmitted to the determined value of the neutron flux. On the contrary, for the determination of the $r \sqrt{\frac{T}{T_0}}$ values only relative measurements of activation rates have to be performed. Therefore, for this application, the knowledge of the composition of the Ag/Al alloy is not necessary, the only important point being the uniformity of the alloy.

10.4 Irradiation of Co/Al and Ag/Al alloys in a neutron flux with known $r \sqrt{\frac{T}{T_0}}$.

Irradiations have been performed in the pneumatic tube PH2 in the graphite reflector very close to the heavy water tank of the Ispra I Reactor. The value of $r \sqrt{\frac{T}{T_0}}$ has been determined by cadmium ratio measurements with gold.

The gold has been irradiated in the form of discs, 2/10 mm thick and 8 mm in diameter, made of dilute alloy of gold and aluminium. (Au content about 0.1%). The uniformity of the

alloy has been tested. Two pairs of irradiations, with and without cadmium, have been performed. For the irradiations under cadmium, capsules of flat cylindrical form with 1 mm thick walls have been utilized. Relative measurements of the gold activation rates have been made by gamma spectroscopy utilizing a 3" x 3" NaI(Tl) crystal connected to a multichannel analyser. The area of the 0.411 KeV gamma photopeak of Au198 has been evaluated.

The activation rates measured in the two pairs of irradiations and the cadmium ratios are reported in Table 10.4.

Table 10.4 : Activation rates and cadmium ratios for gold irradiated in the pneumatic tube PH2.

Pair	Activation rate (arbitrary unit)	Cadmium ratio
1	9.001 (Cd) 1.478	6.09
2	9.075 (Cd) 1.517	5.98

The cadmium ratio values obtained in the two pairs of irradiations are in good agreement.

The calculation of $r\sqrt{\frac{T}{T_0}}$ were made utilizing the average value 6.04. The value of $r\sqrt{\frac{T}{T_0}}$ can be calculated from the value of cadmium ratio using equation [4] reported by W.H. Walker et al. (7).

$$r\sqrt{\frac{T}{T_0}} = \frac{G_{th}(1 - R_{Cd} h)}{(F R_{Cd} - 1) \frac{G_r s_o}{\sigma} + R_{Cd} \left(\frac{1}{K} - w \right)} \quad [4]$$

where

- G_{th} is thermal neutron self shielding factor
- R_{Cd} is the cadmium ratio
- h is the fraction of thermal neutrons transmitted by the cadmium filter
- G_r is the resonance neutron self shielding factor
- F is the transmission of the cadmium filter for neutrons at the resonance energy
- K is a correction factor for the attenuation of the epithermal neutrons in the cadmium filter
- w is a correction factor for the resonance neutrons absorbed by cadmium below the cadmium cut off

- g is a factor to allow for the departure of the cross section from $1/v$ in the thermal region of the neutron spectrum
- s_0 is a factor to allow for the departure of the cross section from $1/v$ in the epithermal region of the neutron spectrum.

In the present experiment the self shielding factors G_{th} and G_r have been assumed to be unity owing to the low percentage of gold in the alloy and the small thickness of the detectors.

The value of h is very small and has been assumed equal to zero: the value of F has been assumed to be unity.

The value of K is reported by C.H. Westcott et al (2): for 1 mm cadmium thickness and isotropic neutron incidence K is equal to 2.29. For W we have assumed the same value utilized by R.G. Hart et al (3) - 0.089 - although the cadmium thickness utilized in that experiment was 0.76 mm in place of 1 mm. In any event, the importance of this factor is small. For s_0 we have taken the value 17.0 and for g the value 1.007 reported in reference (3). With these data and equation (4), we have determined a value of $r \sqrt{\frac{T}{T_0}} = 0.01147$

using a cadmium ratio equal to 6.04 for gold.

In the 2 irradiations without cadmium, we have irradiated, close to the Au/Al alloy foils, wires of Ag/Al alloy (0.1% Ag) and wires of Co/Al alloy (0.1% Co). The activities of these wires can be measured a long time after the irradiation owing to the long half-lives of the isotopes Co60 and Ag110 m. By comparison of the ratio of these activities with the ratio of the activities measured in an unknown spectrum it will be possible to determine the value of $r \sqrt{\frac{T}{T_0}}$ for this spectrum (3).

10.5 Determination of the s_0 value for silver.

The value of s_0 for silver is one of the data required for the determination of $r \sqrt{\frac{T}{T_0}}$ using the procedure reported above. This value has been determined by R.G. Hart et al. (3).

We have made the same measurement as performed by these Authors in order to check the goodness of our measurements and related calculations. For this reason we have irradiated, close to the Au/Al foils, foils of Ag/Al alloy ($\sim 0.1\%$ Ag) and we have determined the cadmium ratio for the nuclear reaction $Ag^{109}(n, \gamma) Ag^{110m}$. The relative activation rates of Ag^{109} have been measured by gamma spectrometry utilizing a 3" x 3" NaI(Tl) crystal connected to a multichannel analyzer.

The activation rates measured in the two pairs of irradiations and the cadmium ratios are reported in table 10.5.

Table 10.5: Activation rates and cadmium ratios for silver irradiated in the pneumatic tube PH 2.

Pair	Activation rate (arbitrary units)	Cadmium ratio
1	49.61 (Cd) 8.769	5.657
2	50.41 (Cd) 8.777	5.743

The cadmium ratios obtained in the two pairs of irradiations are in good agreement.

In the calculation of $s_{o,Ag}$ the average value - 5.70 - was utilized. The values $s_{o,Ag}$ is obtained from cadmium ratio measurements on gold and silver according to equation [5].

$$s_{o,Ag} = \frac{\epsilon_{Ag}}{G_{r,Ag}} \left\{ \frac{Z}{Q} \frac{G_{th,Au}}{G_{th,Ag}} \left[\frac{s_{o,Au} G_{r,Au}}{\epsilon_{Au}} + \frac{1}{F_{Au} K} - \frac{W_{Au}}{F_{Au}} + \frac{H}{F_{Au}} \right] - \left[\frac{1}{F_{Ag} K} - \frac{W_{Ag}}{F_{Ag}} + \frac{H}{F_{Ag}} \right] \right\} \quad [5]$$

$$\text{where } Z = 1 + W_{Ag} \cdot r \sqrt{T/T_0}$$

$$Q = \frac{F_{Ag} R_{Cd,Ag}^{-1}}{F_{Au} R_{Cd,Au}^{-1}}$$

$$H = h \left(r \sqrt{\frac{T}{T_0}} \right)^{-1}$$

The meanings of the terms are the same as for equation [4]. Assuming that $G_{r,Ag}$, $G_{th,Au}$, $G_{th,Ag}$, $G_{r,Au}$, F_{Au} and F_{Ag} are equal to unity and that h is equal to zero, the formula takes the following form.

$$s_{o,Ag} = g_{Ag} \left\{ \frac{Z}{Q} \left[\frac{s_{o,Au}}{g_{Au}} + \frac{1}{K} - W_{Au} \right] - \left[\frac{1}{K} - W_{Ag} \right] \right\}$$

where $Z = 1 + W_{Ag} r \sqrt{T/T_0}$

$$Q = \frac{R_{Cd,Ag}^{-1}}{R_{Cd,Au}^{-1}}$$

The value of $W_{Ag} = 0.098$ and $g_{Ag} = 1.0058$ are taken from reference (3). The value of $r \sqrt{\frac{T}{T_0}}$ has been previously determined by the cadmium ratio measurements on gold and is equal to 0.01147.

Utilizing these data we obtain:

$$Z = 1 + 0.098 \times 0.01147 = 1.0011$$

$$Q = \frac{5.70 - 1}{6.04 - 1} = 0.933$$

$$s_{o,Ag} = 1.0058 \left\{ \frac{1.0011}{0.933} \left[\frac{17.0}{1.007} + \frac{1}{2.29} - 0.089 \right] - \left[\frac{1}{2.29} - 0.098 \right] \right\} = 1.0058 \left\{ 1.073 \times [17.229 - 0.339] \right\} = 18.23$$

We evaluate the total error in the determination as + 2% : hence the final value is 18.23 ± 0.36 . This error does not include the uncertainty in the value of $s_{o,Au}$.

The value is in satisfactory agreement with the value determined by R.G. Hart et al (3) of 18.74 ± 0.08 .

10.6 Design and construction of an automatic system for the scanning of monitor wires.

We plan to insert flux monitor wires on the surface of the UC specimens, for the whole length of the samples, in order to have information on the axial distribution of the neutron flux.

To perform the scanning of the wires, an automatic system has been designed by the Group "Etudes Générales et Génie Radioactif", and is now under assembly. For monitoring the Co/Al and Ag/Al alloys the detector will be a 3" x 3" NaI(Tl) crystal connected to a multichannel analyzer.

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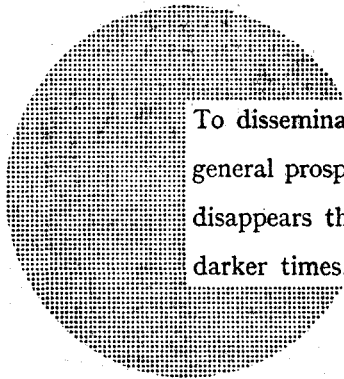
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