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**DETERMINATION OF THE U 235, Pu 239 AND Pu 240
CONTENTS IN MIXED FISSILE MATERIALS
BY MEANS OF ACTIVE AND PASSIVE NEUTRON
TECHNIQUES**

by

G. BIRKHOFF, L. BONDAR and J. LEY

1972



Joint Nuclear Research Centre
Ispra Establishment - Italy

Physics Division

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ABSTRACT

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KEYWORDS

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RAPSODIE
PLUTONIUM
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EFFICIENCY
STATISTICS
URANIUM 235
PLUTONIUM 239
FISSION YIELD
DELAYED NEUTRONS
PROMPT NEUTRONS
NON DESTRUCTIVE TESTING
SPONTANEOUS FISSION
PLUTONIUM 240

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1. INTRODUCTION

The control of fissile material flow during the whole cycle of reactor fuel is becoming a serious problem from safety and fuel management points of view, because of the large expansion of the nuclear industry in the near future. At present, several groups around the world are executing research and development work in this field.

Control systems have been proposed by several control authorities and research teams (1), (2), (3), (4), (5). The common goal is to establish an effective control system with the minimum burden on reactor economy.

An important component of any control system is the nondestructive measurement of fresh fuel in the form of reactor fuel pins, subassembly, and elements. In this paper we investigate a method for measuring the contents of fissile materials (Pu239, U235) of (Pu-U) fuels used in fast breeder reactors, and light water reactors (Pu-recycle). This type of fuel is considered to be of high value for economic and safeguard reasons.

2. METHOD

The method is based on the measurement of prompt and delayed neutron yields of neutron induced fissions. Obviously, the sensitivity of the method is sufficient only if the isotopes to be determined exhibit great differences in the ratios of the prompt to delayed neutron yields per fission. As it concerns the two isotopes of highest interest, U235 and Pu239, the situation is quite favourable in this respect because the delayed neutron fraction of U235 (β_5) is about three times greater than the one of Pu239 (β_9); ($\beta_5=0.0065$, $\beta_9=0.0021$).

In the case of fresh reactor fuel, one has to consider the following isotopes:

- Uranium: U235, U238 (U236 minor isotope)
- Plutonium: Pu239, Pu240 (Pu238, Pu241 minor isotopes).

The minor isotopes (U236, Pu238 and Pu241) can be usually neglected due to their much lower concentrations.

Discrimination against U238 can be obtained by irradiating the sample in a field of neutrons with a spectrum below the fission threshold of this isotope ($E_n < 0.8$ MeV). Pu240 can be determined by measuring the spontaneous fission neutron emission^{(6), (7)}. The remaining two isotopes U235 and Pu239 are determinable from the prompt and delayed fission neutron yields.

3. BASIC EQUATIONS

The (U-Pu) sample is exposed to a neutron flux $\phi_0(\vec{r}, E)$ which is generated by a neutron source. Due to interaction with the sample (scattering, absorption, production) the neutron flux is perturbed.

$$\phi_0(\vec{r}, E) \rightarrow \phi(\vec{r}, E)$$

The production (P_i) of fission neutrons from an isotope i of the sample amounts to:

$$(1) \quad P_i = \int_V \int_E N(\vec{r})_i \nu(E)_i \sigma_f(E)_i \phi(\vec{r}, E) dE dV = (N \nu \sigma_f)_i \phi \cdot V$$

with

$$(1a) \quad \phi = \frac{1}{V} \int_V \int_E \phi(\vec{r}, E) dE dV$$

$$(1b) \quad (N \nu \sigma_f)_i = (\nu \Sigma_f)_i = \frac{\int_V \int_E N(\vec{r})_i \nu(E)_i \sigma_f(E)_i \phi(\vec{r}, E) dE dV}{\int_V \int_E \phi(\vec{r}, E) dE dV}$$

If $\varepsilon(\vec{r})_i$ is the detection probability of a fission neutron emitted by an isotope i born in a volume element dV at \vec{r} the detector counting rate is given by the following expression:

$$(2) \quad C_i = \int_V \int_E \varepsilon(\vec{r})_i N(\vec{r})_i \nu(E)_i \sigma_f(E)_i \phi(\vec{r}, E) dE dV$$

$$(2a) \quad = \varepsilon_i (N \nu \sigma_f)_i \phi V$$

with

$$(2b) \quad \varepsilon_i = \frac{\int_V \int_E \varepsilon(\vec{r})_i N(\vec{r})_i \nu(E)_i \sigma_f(E)_i \phi(\vec{r}, E) dE dV}{\int_V \int_E N(\vec{r})_i \nu(E)_i \sigma_f(E)_i \phi(\vec{r}, E) dE dV}$$

The counting rate of delayed neutrons, C_i^d , which are measured with a detection probability $\epsilon_i^d(\vec{r})$, is:

$$(3) \quad C_i^d = \int_V \int_E \epsilon_i^d(\vec{r}) \beta_i \cdot N(\vec{r})_i \nu(E)_i \sigma_f(E)_i \phi(\vec{r}, E) dE dV =$$

$$= \epsilon_i^d \beta_i (N \nu \sigma_f)_i \phi \cdot V$$

with

$$(3a) \quad \epsilon_i^d = \frac{\int_V \int_E \epsilon_i^d(\vec{r}) \cdot N(\vec{r})_i \nu(E)_i \sigma_f(E)_i \phi(\vec{r}, E) dE dV}{\int_V \int_E N(\vec{r})_i \nu(E)_i \sigma_f(E)_i \phi(\vec{r}, E) dE dV}$$

(β_i = delayed neutron fraction of fission neutrons from isotope i)

The respective counting rates of fission neutrons emitted by all isotopes are:

$$(4) \quad C = \sum_i C_i = \sum_i \epsilon_i (N \nu \sigma_f)_i \phi \cdot V$$

and

$$(5) \quad C^d = \sum_i C_i^d = \sum_i \epsilon_i^d \beta_i (N \nu \sigma_f)_i \phi \cdot V$$

In principle it is possible to determine experimentally C , C^d , ϕ_s (surface flux at the sample) V , β_i and the detection probabilities ϵ_i and ϵ_i^d of neutrons emitted from the surface of the sample. On the contrary, one can not measure the flux distribution $\phi(\vec{r}, E)$, by a non destructive technique, inside the sample because it is depending on its nuclear composition. One can therefore only determine from experimental quantities averaged concentrations of maximum two isotopes weighted by not well known distribution functions of flux, fission cross section and detection probabilities. The situation may be improved essentially by establishing the following experimental conditions:

- (a) spectrum of source neutron flux in the sample below fission threshold of U238 and other fast fission isotopes (sub MeV flux),
- (b) slight flux perturbation by the sample itself,

(c) spatial uniformity of flux and detection probabilities.

Under these conditions one may assume that fission neutrons are produced uniformly throughout the sample by the sub-MeV source neutrons and that they are only slightly multiplied by secondary fissions. If the contributions due to secondary fission and due to minor isotopes (U236, Pu241 and Pu240) are neglected, one obtains the following equations:

$$(6) \quad C = C_5 + C_9$$

$$(7) \quad C^d = C_5^d + C_9^d$$

$$(8) \quad C_5 = \epsilon N_5 (\nu \sigma_f)_5 \phi V$$

$$(9) \quad C_9 = \epsilon N_9 (\nu \sigma_f)_9 \phi V$$

$$(10) \quad C_5^d = \epsilon^d \cdot \beta_5 (\nu \sigma_f)_5 \phi V N_5$$

$$(11) \quad C_9^d = \epsilon^d \cdot \beta_9 (\nu \sigma_f)_9 \phi V N_9$$

with

$$(12) \quad N_i = \frac{1}{V} \int_V N(\vec{r})_i dV$$

$$(13) \quad \phi = \int_E \phi(E) dE$$

$$(\nu \sigma_f)_i = \frac{\int_E \nu(E)_i \sigma_f(E)_i \phi(E) dE}{\int_E \phi(E) dE}$$

resolving eq. (6), (7), ..., (11) it follows:

$$(14) \quad N_5 \cdot V = \frac{C - \epsilon / \epsilon^d \cdot C^d / \beta_9}{\phi (\nu \sigma_f)_5 \epsilon (1 - \beta_5 / \beta_9)} = A(C - k_5 \cdot C^d)$$

with

$$(14a) \quad A = \frac{1}{\phi (\nu \sigma_f)_5 (1 - \beta_5 / \beta_9) \cdot \epsilon}$$

$$(14b) \quad k_5 = \epsilon / \epsilon^d \cdot 1 / \beta_9$$

$$(15) \quad N_9 \cdot V = \frac{C - \varepsilon/\varepsilon^d \cdot C^d/\beta_5}{\varphi(\nu\sigma_f)_9 \varepsilon(1-\beta_9/\beta_5)} = B(C - k_9 C^d)$$

with

$$(15a) \quad B = \frac{1}{\varphi(\nu\sigma_f)_9 \varepsilon(1-\beta_9/\beta_5)}$$

$$(15b) \quad k_9 = \varepsilon/\varepsilon^d \cdot 1/\beta_5$$

The four constants A, B, k_5 , k_9 can be determined by calibration measurements with samples of various concentrations of N_5 and N_9 .

As concerns the contributions due to secondary fission and minor isotopes, they may be treated as second order effects theoretically.

4. OPTIMIZATION STUDIES

The goal of the optimization studies is to approach closely the assumption of the eq. (14) and (15) of the previous section. Moreover, the costs of a measurement should be minimized. The last condition can be subdivided into minimization of:

- source neutrons costs,
- apparatus costs,
- operational costs,
- and maximization of source neutron flux per unit source inside the sample.

Obviously, the maximum source neutron flux in the sample is obtained by inserting the sample into a channel or chamber inside the neutron source itself. The choice of components of which the apparatus is built up, will be subjected to all the mentioned optimization criteria whenever possible.

4.1 Choice of Fuel Type

The starting point of any optimization study is the definition of the geometry- and isotopic composition-ranges of the fuel samples to be measured.

In the present paper as an example of the proposed method, the measurement of fuel from the French fast breeder reactor RAPSODIE⁽⁸⁾⁽¹¹⁾ is considered.

This case has been chosen because there is already a considerable production of this fuel and a request of control instruments. Once the method has been fully tested, the measurement of other (Pu-U) fuel would be almost a matter of designing a special apparatus. The geometrical and isotopic data of the RAPSODIE fuel are listed in Table I.

4.2 Choice of Neutron Source

Most of the available neutron sources produce neutrons with energies above the fission threshold of U238. In these cases the neutron spectra must be transferred into the sub-MeV region by a proper spectrum shifter. On the other side, the neutron energy should be higher than about 1 KeV because of the high absorption cross section of the (U-Pu) sample in the sub-KeV region, which causes a significant self shielding. Extensive studies of such spectrum shifters have been done by the LOS ALAMOS group of KEEPIN for a 14 MeV and a fission spectrum source⁽⁹⁾. The results have shown that it is quite possible to shift about 85% of all source neutrons below the U238 fission threshold. Nevertheless, the remaining 15% of source neutrons, above the threshold, create serious and quite unresolvable problems for the analysis of the envisaged irradiation experiment with a (U-Pu) sample. For these reasons, sources with spectra above the U238 fission threshold have been ruled out. Among the sub-MeV neutron sources there are isotopic and accelerator sources. The more practical sources of this kind are summarized in Table II.

Isotopic sources have a great advantage due to the stability of neutron emission rate and spectrum. No operational staff is required and measurements can be easily fully automatized. As concerns the neutron yield, accelerators are much more powerfull. Moreover, accelerators have the advantage of being variable, with respect to neutron energy and yield.

At present, all types of accelerator sub-MeV neutron sources are quite

costly. It seems that MICROTRONS in connection with D_2O targets have a good chance in the future, for being the most adapted neutron sources. Isotopic (α, n) sources with the required intensity (10^9 n/sec) are actually not available for various reasons. There is no hope that sources of more than 10^8 n/sec will be produced in the near future for less than \$ 100,000. --. Sb-Be (γ, n) sources with a neutron output of about 10^{10} n/sec have already been built in some laboratories. The production of Sb124 with the required intensity in an isotopes production reactor, is generally quite expensive. Moreover, due to the reduced market of such items, it is hard to get a good estimation of its mean price. The price we give in Table II is based on an offer from the reactor SORIN, located at SALUGGIA (7 MW Swimming Pool Reactor, Owner: FIAT). Nevertheless, if one would verify an increased demand, a significant drop of the price might be expected. The main drawback of the Sb-Be source is the relatively short half-life which charges the users with the organization of periodic reactivation of the Sb124.

Taking into account all aspects, it is felt that the choice of the Sb124-Be source is the most feasible solution at present.

4.3 Source-Sample Geometry

The source sample geometry is chosen as:

- (a) to approach as closely as possible the assumption of eq. (14) and (15), i. e.
 - spatial uniformity of source neutron flux,
 - minimum of spectrum perturbation by shielding materials in the surroundings;
- (b) to achieve maximum source neutron flux in the fuel sample.

In general it is necessary to surround the source by shielding material against γ -radiation, as lead (Pb), tungsten (W) etc., and against neutron radiation, as paraffin etc. $(CH_2)_n$.

The schematic of an irradiation chamber consisting of an Sb-Be source, surrounded by Pb and $(CH_2)_n$ is shown in Fig. 1. The source has the form of

a hollow cylinder and the fuel sample is inserted for irradiation into the central channel of it, where the neutron flux is spatially flat due to a uniform distribution of γ -activity of the Sb-cylinder. Obviously, this geometry is the optimum as concerns the flux level in the fuel sample. In cases where the fuel pin is longer than the neutron source, axial flux uniformity can be achieved by oscillating the fuel rod or the source.

It is clear that the flux shape can be deformed considerably by the fuel sample. In radial direction there will be always a certain flux depression which depends strongly on the neutron spectrum, the diameter of the fuel rod and its absorption properties. Axial heterogeneity of the fuel rod may cause a deformation of the axial flux shape. Obviously, the undesired flux deformations are the stronger, the softer the neutron spectrum and the bigger the diameter of the fuel rod are. From all those considerations we may immediately conclude that the method is particularly adapted for small diameter fuel rods, i. e. fuel pins. This case is especially envisaged for application.

4.4 Neutron Spectrum Inside a Fuel Pin During Irradiation

The neutron spectrum inside a fuel pin located in the central channel of the neutron source was calculated by means of the Monte Carlo transport theory (TIMOC code)⁽¹⁰⁾. Particularly the influences of shielding materials (Pb, CH₂) have been investigated. Results of those calculations are summarized in Table III.

The given geometrical data refer to the scheme of fig. 1. The wall thickness of the Sb-tube (0.6 cm) is chosen as to achieve a specific activity of 15 Ci per cm length of tube after an irradiation time of 3 weeks in a thermal neutron flux of 3×10^{13} n/cm².sec. (Routine run of the ISPRA-I reactor). Surrounding the Sb-tube by a Be-tube of 1.5 cm wall thickness, the specific neutron source strength amounts to about 10^8 n/cm² per cm length of the source cylinder. The neutron flux in the central channel of the source is of about 5×10^7 n/cm².sec., which gives a sufficient fission rate in the fuel rod. The boron tube around the fuel rod shields against moderated neutrons.

The results of the spectrum calculation can be summarized as follows:

- the unperturbed source neutron spectrum (i. e. without any shielding around) lies between about 1 KeV and the starting energy of 26 KeV (slowing down due to beryllium),
- significant spectrum broadening appears if the source is surrounded by dense hydrogenous materials at distances less than about 10 cm from the source,
- lead around the source broadens the neutron spectrum only slightly but raises the flux in the central channel by a factor of about 2 due to neutron reflection,
- increasing the wall thickness of the Be-tube produces spectrum broadening and a decrease of flux per unit source.

Taking into account the shielding requirements we may draw from these results the following conclusions:

- the neutron source should be surrounded by successive layers of lead (Pb) (or other heavy materials) and paraffine $(CH_2)_n$. The distance between source surface and hydrogenous materials should be greater than 10 cm. In the following chapters we consider always a Pb-layer of 20 cm thickness followed by a $(CH_2)_n$ layer of 10 cm thickness.

4.5 Flux Perturbation Due to the Fuel Sample

Insertion of a fuel sample into the source channel causes perturbations of the neutron spectrum due to scattering absorption and production of neutrons. Obviously, the perturbation increases with the mass of fuel material inserted. Two extreme cases of a RAPSODIE fuel were investigated:

1. fuel pin of 0.7 cm diameter,
2. fuel rod of 6 cm diameter (equivalent to a fuel element bundle of 73 fuel pins)

The results of the spectrum calculations are shown in fig. 2. In fig. 3

and fig. 4 the radial flux distributions are plotted in a two-group representation ($\phi_1: E_n \geq 0.8 \text{ MeV}$, $\phi_2: E_n < 0.8 \text{ MeV}$, $\phi = \phi_1 + \phi_2$).

The results are summarized as follows:

- (a) insertion of a small diameter fuel pin ($d = 0.7 \text{ cm}$) perturbs the neutron spectrum very little. The radial flux distribution is rather flat.
- (b) insertion of a big diameter fuel rod ($d = 6 \text{ cm}$) causes strong spectral perturbation due to absorption of source neutrons and high fission neutron production. The depression of the sub-MeV neutron flux ($\phi_{\text{average}}/\phi_{\text{surface}}$) is about 0.9 and of the above MeV flux about 1.2.

5. DETECTION OF FISSION NEUTRONS

The detection system has to measure:

- 1. fission neutrons during irradiation with source neutrons,
- 2. delayed neutrons from neutron induced fission,
- 3. spontaneous fission neutrons.

According to the assumption of section 3, the detection probabilities of neutrons within the three categories should be independent of the emission point and the emitter itself.

These requirements have a geometrical and a spectral aspect. From geometrical point of view, the problem can be resolved quite easily by establishing a so-called "good geometry" (each volume element of the fuel sample sees the detector under the same solid angle). The spectral aspect means that a neutron detector response generally depends on the neutron energy spectrum, which is affected by the material to be passed through by the neutrons before reaching the detector. A "good spectrum" is obtained if the average spectrum of fission neutrons at the detector location does not depend on the emission point.

This condition can be verified by placing heavy scattering material (Pb, W) between the fuel sample and the detector in such a manner that the neu-

tron spectrum at the detector position is essentially determined by the scatterer. As concerns the primary fission neutron spectrum, one has to consider prompt and delayed neutrons. As far as known, no measurable difference exists in prompt neutron spectra of all fissionable isotopes (spontaneous or neutron induced fissions). Delayed neutron spectra show significant differences from isotope to isotope but in the case of interest, i.e. U235 and Pu239, the differences are very small.

5.1 Detection of Fission Neutrons During Irradiation

During irradiation of the fuel sample the detector is immersed in a field of all kinds of fission neutrons and in addition of neutrons and γ -rays from the source. The detector must be able to discriminate against the source neutrons and γ -rays. It should further be sufficiently sensitive for the fission neutrons and convenient in operation. Taking account of these requirements, we consider He3 (n, p)H3 and proton and He4 recoil proportional counters in connection with pulse height discrimination.

Properties of these detectors are given in Table IV. Care must be taken for a proper attenuation of the low energy neutron and γ -fluxes at the counter position. This can be achieved by shielding the counter by tungsten (W), which has a very strong γ -ray attenuation and moreover strong neutron resonance absorption in the sub-KeV region. Boron carbide (B_4C) should be used further for attenuation of (eV)-neutrons.

5.2 Detection of Delayed Neutrons

Delayed neutrons can be easily measured after switching off the neutron source. The persisting neutron flux is composed of delayed neutrons, spontaneous fission neutrons and neutrons from (α, n) reactions. Because of the strong decrease of the persisting neutron flux, the fuel sample should be transferred immediately after the irradiation into a high efficiency neutron counter assembly. This assembly is usually built up from He3 or BF3 counters which are embedded in a neutron moderator (4π counter). It can be constructed in such a manner that it is especially highly sensitive for delayed

neutrons which have an average neutron energy of about 0.5 MeV and less sensitivity for the spontaneous fission neutrons and neutrons from (α, n) reactions which have a much higher mean energy. Separation of delayed neutrons is easily obtainable due to their time behaviour (decay).

5.3 Detection of Spontaneous Fission Neutrons

After the complete decay of the delayed neutrons, only spontaneous fission neutrons and neutrons from (α, n) reactions remain and can be measured with the same counter. Due to the multiple neutron emission by spontaneous fission events, the detector signals exhibit a certain time correlation which can be used for the discrimination against neutrons from single neutron emission events ((α, n) reactions)⁽⁷⁾.

6. CONCLUSION

The optimization studies theoretically demonstrate the feasibility of the proposed method. It is clear, however, that in principle one can falsificate a measurement for instance by heterogeneous distribution of fissile material and addition of a moderator in such a manner that the weight of the rod and the neutronic signal will correspond to a calibration standard of the same shape but with a different fissile material content.

If the measurement should be tamperproof, as required for control purposes, only small diameter fuel pins must be regarded where falsification of any kind is very unlikely, contrary to big size samples. If, however, problems of tamperproofness are unimportant, as in cases of quality control of a fabrication plant, or a power reactor station, the method should be applicable too for fuel elements (equivalent to big diameter rod). The sensitivity and accuracy of a measurement depend on many experimental factors and must therefore be determined experimentally. An estimation will be given in connection with the description of an apparatus in the following section.

7. APPARATUS

The lay out of the apparatus is generally based on the results of the preceding theoretical studies. Its main purpose is to demonstrate experimentally the feasibility of the method. The guide lines of the technical design are simplicity and flexibility. The whole apparatus might be subdivided in three main units: irradiation chamber with detection system for fission neutrons; high efficiency counter for delayed neutrons and spontaneous fission neutrons measurement; pneumatic system for switching on and off the (Sb-Be) neutron source, movement of the Be-cylinder and fuel pin transfer.

The irradiation chamber serves for two purposes: it is used as lead castle for the transport and storage of the Sb^{124} source and for the irradiation of the fuel pins. Mounting and dismounting of the three units can be performed rather easily and remotely in order to minimize radiation hazards. A semi-schematic view of the mechanical part of the apparatus is shown in fig. 5.

The functioning of this apparatus can be easily understood from the description of the measuring cycle which is the following:

1. insertion of the fuel pin into the irradiation chamber,
2. switching on of neutron source by drive-in of Be-cylinder,
3. measurement of fission neutrons by a biased He^3 (or proton or helium recoil) proportional counter (5 min.),
4. shooting out of Be-cylinder for switching off the neutron source and rapid transfer of fuel pin into the high efficiency counter for measurement of delayed and spontaneous fission neutrons (10 min.).

For the automatic control and performance of the measurements, conventional electronics can be utilized.

The most interesting characteristical data, calculated by the TIMOC code are listed below⁽¹⁰⁾:

- | | |
|---------------------------------|--|
| - neutron source strength: | 2.10^9 n/sec. |
| - neutron flux in the fuel pin: | $\phi_1(E > 0.8 \text{ MeV}) = 2.0 \cdot 10^6 \text{ n/cm}^2 \cdot \text{sec}$ |

$$\phi_2(E_n < 0.8 \text{ MeV}) = 5.6 \cdot 10^7 \text{ n/cm}^2 \cdot \text{sec.}$$

- production of neutrons per unit volume of fuel pin:

isotope	<u>prompt neutrons</u>	<u>delayed neutrons in equilibrium</u>
U235	$4.50 \cdot 10^6 \text{ n/cm}^3 \cdot \text{sec.}$	$2.84 \cdot 10^4 \text{ n/cm}^3 \cdot \text{sec.}$
U238	$1.85 \cdot 10^4 \text{ "}$	$2.96 \cdot 10^2 \text{ "}$
Pu239	$1.93 \cdot 10^6 \text{ "}$	$4.04 \cdot 10^3 \text{ "}$
Pu240	$1.34 \cdot 10^4 \text{ "}$	$3.50 \cdot 10^1 \text{ "}$

- fission neutron flux in the counter channel: $\phi(E_n > 0.4 \text{ MeV}) = 1.48 \cdot 10^4 \text{ n/cm}^2 \cdot \text{sec.}$

The accuracy of the measurements is estimated on the following assumptions.

Using four He3 counters of 2.5 cm diameter and 50 cm length with 6 atm of pressure as fission neutron detector (bias - 0.5 MeV), the total counting rate amounts to 600 cps corresponding to a counting efficiency of $2.5 \cdot 10^{-5}$.

In principle it would be possible to raise considerably the counting rate by increasing the He3 pressure, but as the rate of sub-threshold pulses will be almost 100 times higher, serious problems of pulse pile-up will arise. No difficulties are expected from the γ -radiation due to the effective shielding by the tungsten slab. A similar fission neutron counting rate can be achieved with a proton recoil counter of 5 cm diameter and 50 cm length filled with 1 atm of H1. In this case problems of pulse pile-up due to sub-threshold neutrons are very reduced but this advantage is partially nullified by γ -pulse pile-up problems. Using a 2 atm filling of H1 the counting rate of fission neutrons amounts to about 1200 cps ($\epsilon \sim 5 \cdot 10^{-5}$). The counting efficiency can be still increased by a high pressure He4 recoil counter. The advantages of a He4 recoil counter with respect to a proton recoil counter are the higher scattering cross section and the strong forward peaking of He4 recoils for MeV neutrons (resonance of 7 barn at 1 MeV). These features result in a high counting efficiency of fission neutrons and a very strong discrimination of sub-MeV neutrons. A disadvantage is the smaller neutron energy transfer.

It is foreseen to investigate experimentally all these counter types (He3, proton and He4 recoil) in order to find out the best solution. From the estimation it is thought that it will be possible to measure the rate of fission neutrons with a statistical accuracy of 0.3% within 5 min. of counting time. The delayed neutrons are measured with an efficiency (ϵ^d) of 0.3. Assuming a delay time of 0.5 sec between the end of irradiation and the start of measurement and a counting time of 5 min., the total count of delayed neutrons amounts to $5 \cdot 10^5$.

The spontaneous fission neutrons are counted with an efficiency of about 0.2 corresponding to a counting rate of 320 cps and a total of $9.6 \cdot 10^4$ counts during the 5 min. measuring time. The statistical accuracy of the delayed neutron measurement amounts therefore to about 0.2%. Under these conditions the statistical error of the concentrations of U235 and Pu239 becomes 0.5% and 1.7% respectively, according to eq. 1 and 13. The uncertainties due to the calibration should be of the same order of magnitude. Thus, the overall accuracy is estimated to be about $\pm 1\%$ for N_5 and $\pm 3.5\%$ for N_9 .

From the coincidence analysis⁽⁶⁾ of the pulses after the decay of delayed neutrons, the concentration of Pu240 can be determined with a statistical accuracy of $\pm 3\%$ within a 5 min. measuring time. A further information on the concentration of Pu239 can be obtained from the measurement of the γ -rays emitted by this isotope.

Combining all measurements of Pu-isotopes it seems possible to determine the concentration of Pu239 with an accuracy of $\pm 2\%$.

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TABLE 1 - CHARACTERISTICS OF RAPSODIE FUEL

Mass and isotopic compositions:

PuO₂ in (U, Pu)O₂ 25.91%

Pu239 89.0%

U234 0.37%

Pu240 9.7%

U235 60.15%

Pu241 1.2%

U236 0.31%

Pu242 0.07%

U238 39.17%

Geometrical data:

pellets;	real density	10.6 gr/cm ³
	diameter	5.57 mm
	height	10.0 mm
pin;	total length	485 mm
	active length	340 mm
	cladding OD	6.70 mm SS 316L or
	ID	5.80 mm Z3 CND 18-12
	total mass of oxide	87.3 gr
bundle;	geometrical shape:	hexagonal
	number of fuel pins:	37

TABLE II - ISOTOPIC AND ACCELERATORS SUB MeV NEUTRON SOURCES

Accelerator Sources				Isotopic Sources		
Characteristics	Microtron	Van de Graaf	Lineac	Sb124-Be	Po210-Li7	Am241-Li7
Energy (MeV): e-beam current: pulse length: repetition rate:	4-8 50-200 mA(p) 2 μ sec	2-3 1 mA pulsed or continuous	4-8 50-200 mA(q) 2 μ sec \leq 200	Reaction γ -n Emitter Sb124 T 1/2 60.9 d n-energy 0.026 MeV	α -n Po210 138.5 d < 1 MeV	α -n Am241 462 y < 1 MeV
Price	~\$ 100,000	~\$ 150,000	~\$ 150,000	< \$ 2,000		
Maintenance costs	~\$/y 15,000	~\$/y 20,000	~\$/y 20,000	-	-	-
Peripheral support equipment	-	~\$ 50,000	~\$ 20,000	activation for 300 Ci \$ 600	-	-
Staff	1 techn/+mn	1 techn/+mn	1 techn/+mn	-	-	-
Ease of operation	yes	no	yes	yes	yes	yes
Portability	"	"	"	"	"	"
n-spectrum stability	good	good	good	perfect	perfect	perfect
Flexibility	applicable for γ -n inter- rogation with va- riable spectra and intensities	variable n- spectra and intensities	as microtron	limited varia- tion of n-spec- trum and inten- sity	no	no
Reliability	high	medium	high	perfect	perfect	perfect

continued TABLE II

Accelerator Sources				Isotopic Sources		
Characteristics	Microtron	Van der Graaf	Lineac	Sb124-Be	Po210-Li7	Am241-Li7
Factor of charge	~80%	~ 60%	~ 80%	100%	100%	100%
Max. n-output	$10^{13}/10^{14}$ (γ -n;pulsed operation)	$\sim 10^{11} [4^7(p, n);$ T(p, n)]	$10^{13}/10^{14}$ (γ -n; pulsed operation)	$\sim 6 \cdot 10^6$ n/sec/ Ci	$1.7 \cdot 10^5$ n/sec/ Ci	$4 \cdot 10^4$ n/sec/ Ci
Shielding	compact	bulky	bulky	compact	compact	compact

TABLE II - NEUTRON SPECTRUM $\overline{\phi(E)}$ INSIDE A NEUTRON-IRRADIATED (U-Pu)O₂ FUEL PIN, FOR
GEOMETRY SEE FIG. 1

E_n [KeV]	$\overline{\phi(E)}$ [10^{-8} neutron/cm ² .sec] per source neutron						$\Sigma_a(E)$ [cm ⁻¹]
	1 cm Be					3 cm Be	
	0 cm void	10 cm void	20 cm void	40 cm void	20 cm Pb	20 cm Pb	
0 - .000025	-	-	-	-	-	-	-
25 - .000215	-	-	-	-	-	-	-
215 - .000465	-	-	-	-	-	3460.	-
465 - .00100	2920.	2160.	-	-	-	-	2.48
100 - .00215	993.	-	-	-	-	347.	-
215 - .00465	893.	-	-	-	-	-	.368
465 - .0100	986.	22.3	-	-	12.9	-	2.48
100 - .0215	-	-	-	-	-	-	-
215 - .0465	69.9	-	-	-	-	43.8	1.53
465 - .100	432.	-	-	-	-	347.	1.32
100 - .215	207.	-	-	-	-	248.	.687
.215 - .465	71.5	6.63	1.98	-	4.62	109.	.431
.465 - 1.00	79.1	1.54	1.61	1.61	33.2	196.	.283
1.00 - 2.15	19.8	2.86	3.09	2.94	35.5	173.	.173
2.15 - 4.65	27.6	8.69	7.49	8.05	57.3	84.0	.126
4.65 - 10.0	27.6	21.9	22.1	21.9	70.5	60.1	.0965
10.0 - 21.5	49.2	45.6	46.4	45.6	95.7	35.5	.0763
21.5 - 46.5	24.7	24.2	24.9	24.5	34.9	7.95	.0583
46.5 - 100.	.0867	.0236	.0239	.0237	.0622	-	.0439
100. - 200.	.00139	.00137	.00106	.00094	-	.0398	.0347
200. - 400.	.0698	.0143	.0309	.0204	-	.0420	.0290
400. - 800.	.0690	.0313	.0247	.0221	.163	.0944	.0261
800. - 1400.	.0251	.0183	.0178	.0236	.0601	.0297	.0269
1400. - 2500.	.0344	.0242	.0177	.0188	.0444	.0255	.0300
2500. - 4000.	.0182	.00726	.0110	.00725	.0209	.0127	.0300
4000. - 6500.	.00363	.00099	.00432	.00121	.0123	.00212	.0314
6500. - 10500.	.00027	.00012	.00021	.0012	.00642	.00152	.0500

TABLE IV - PROPERTIES OF He3, PROTON AND He4 RECOIL COUNTERS

REACTION		CROSS SECTION (in barns)				ENERGY OF EMITTED PARTICLES (in MeV)			
He3	He3(n, p)H3, Q=0.764	neutron energy				neutron energy			
		0.025 eV	0.5 MeV	1 MeV	2 MeV	0.025 eV	0.5 MeV	1 MeV	2 MeV
		5.040	0.74	0.72	0.72	H1 0.573	0.948	1.323	2.073
		5.441	0.91	0.81	0.82	H3 0.191	0.316	0.441	0.691
						0.764	1.264	1.764	2.764
H1	(n, p) scattering spherical symmetry up to $E_n = 14 \text{ MeV}$	-	6.5	4.5	3	-	≤ 0.5	≤ 1.00	≤ 2.00
He4	(n, He4) scattering	-	1.4	6	4	-	≤ 0.32	≤ 0.64	≤ 1.28
		strong forward peaking of He4 recoils							
		resonance at $\sim 1 \text{ MeV}$ rapidly decreasing below this energy.							

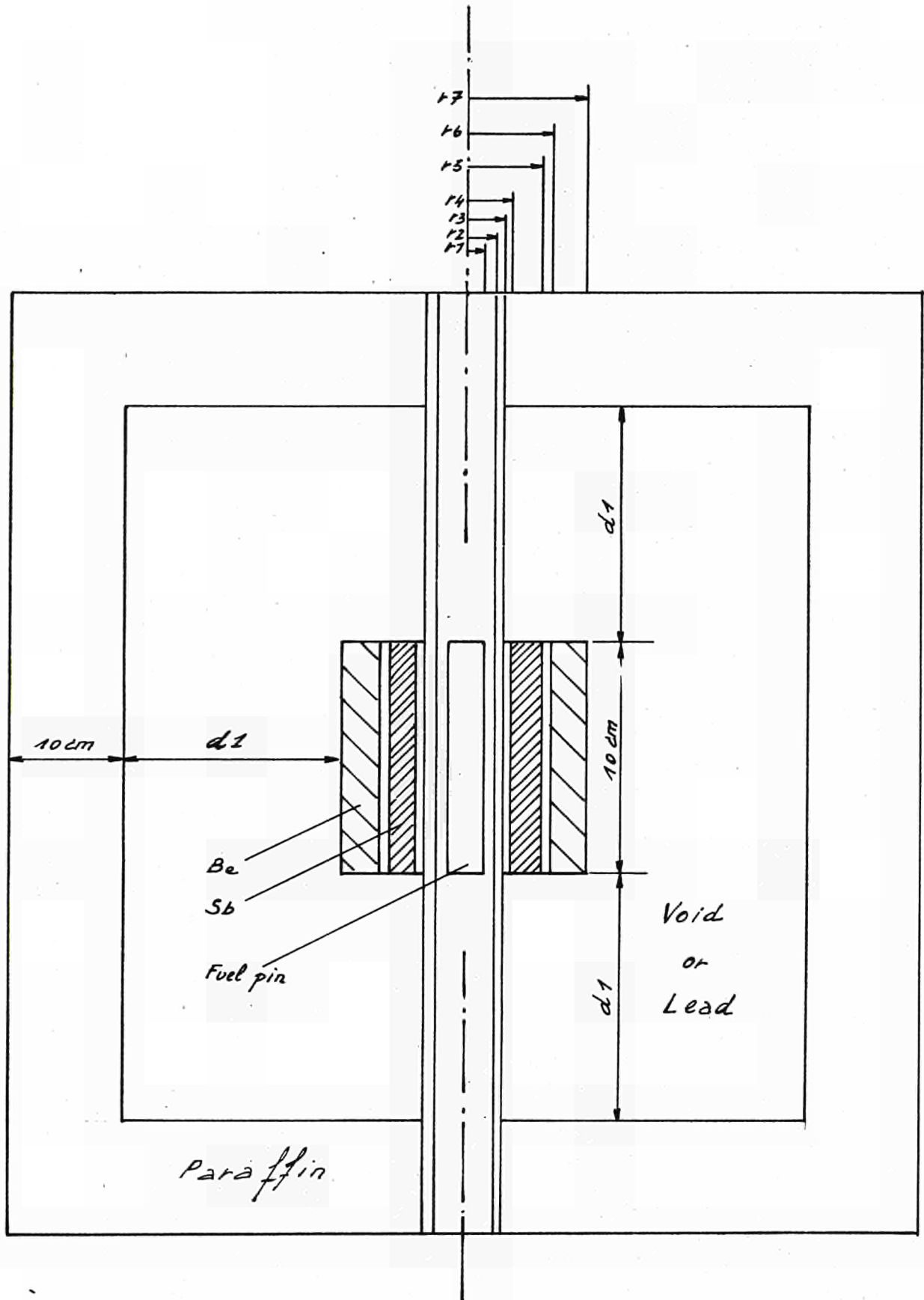
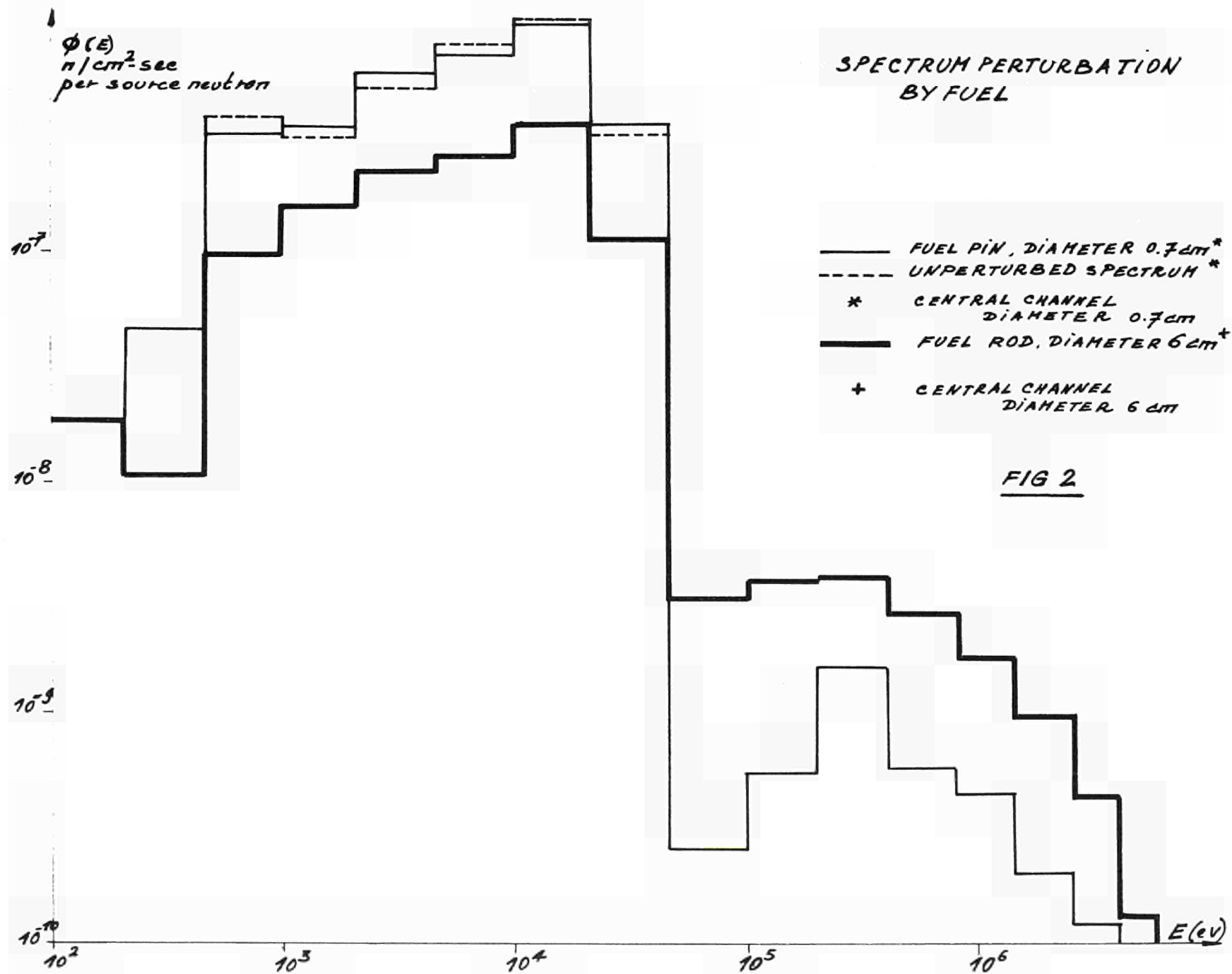
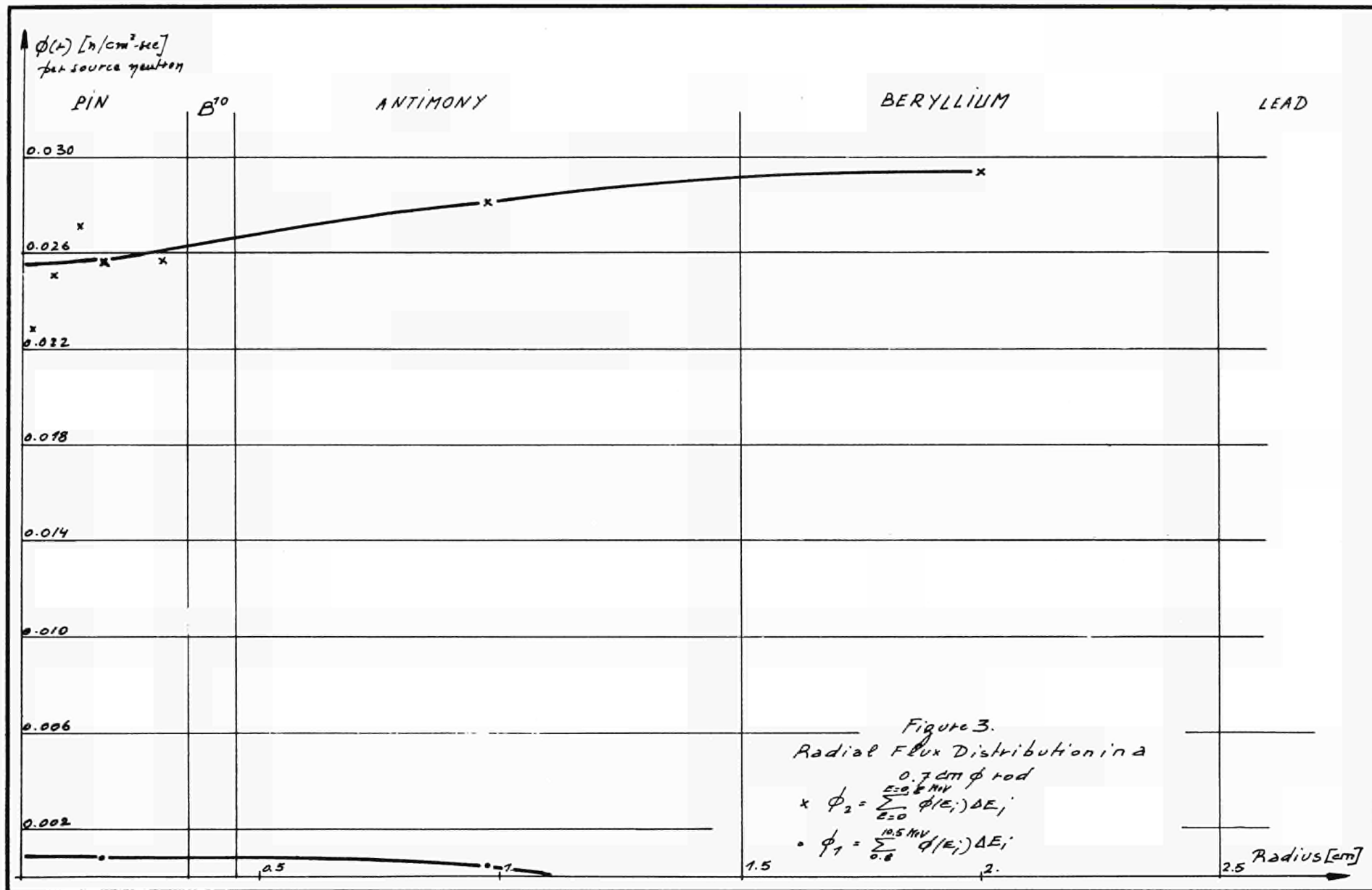
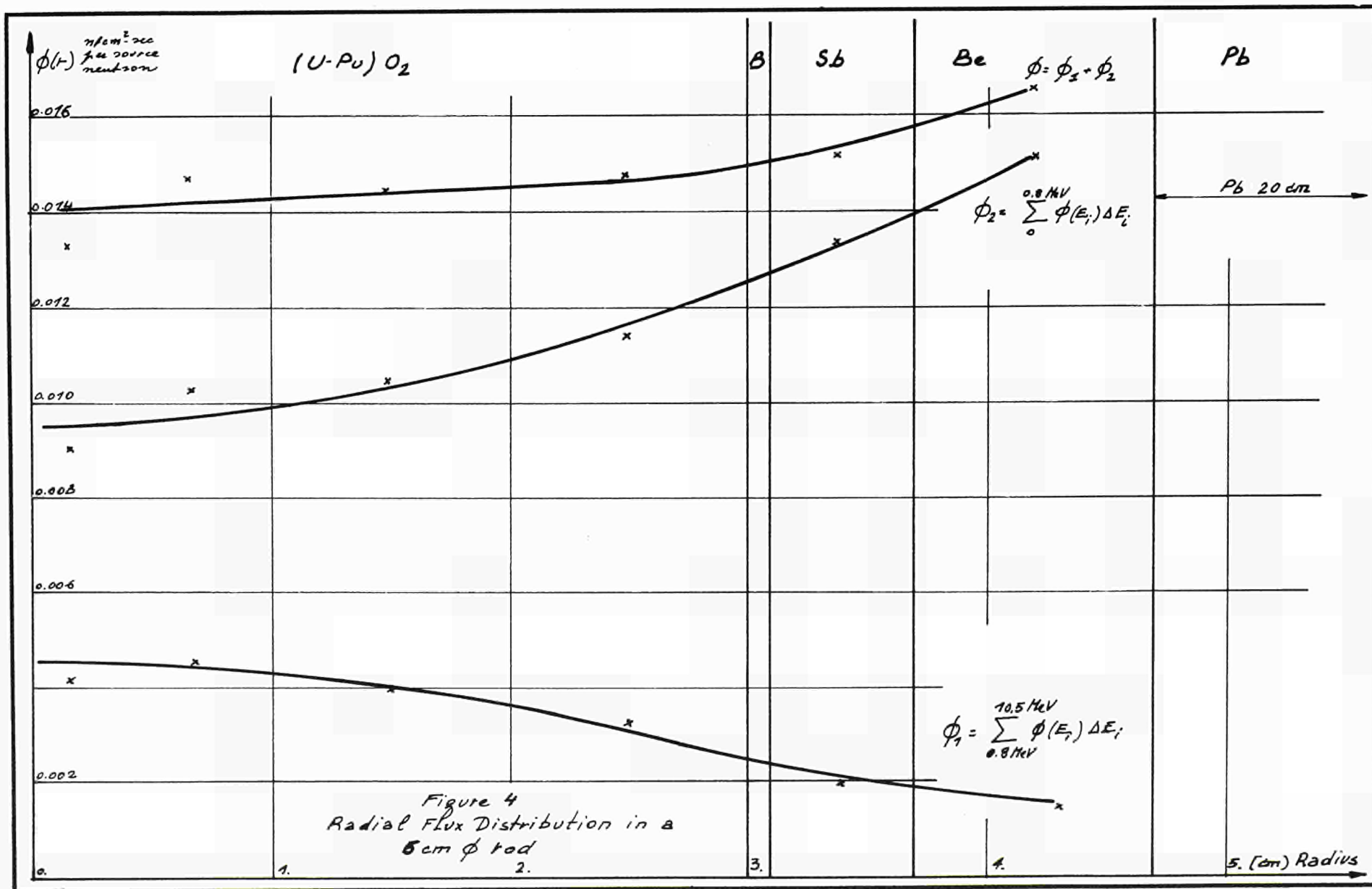


Fig.1 Irradiation Chamber (scheme)



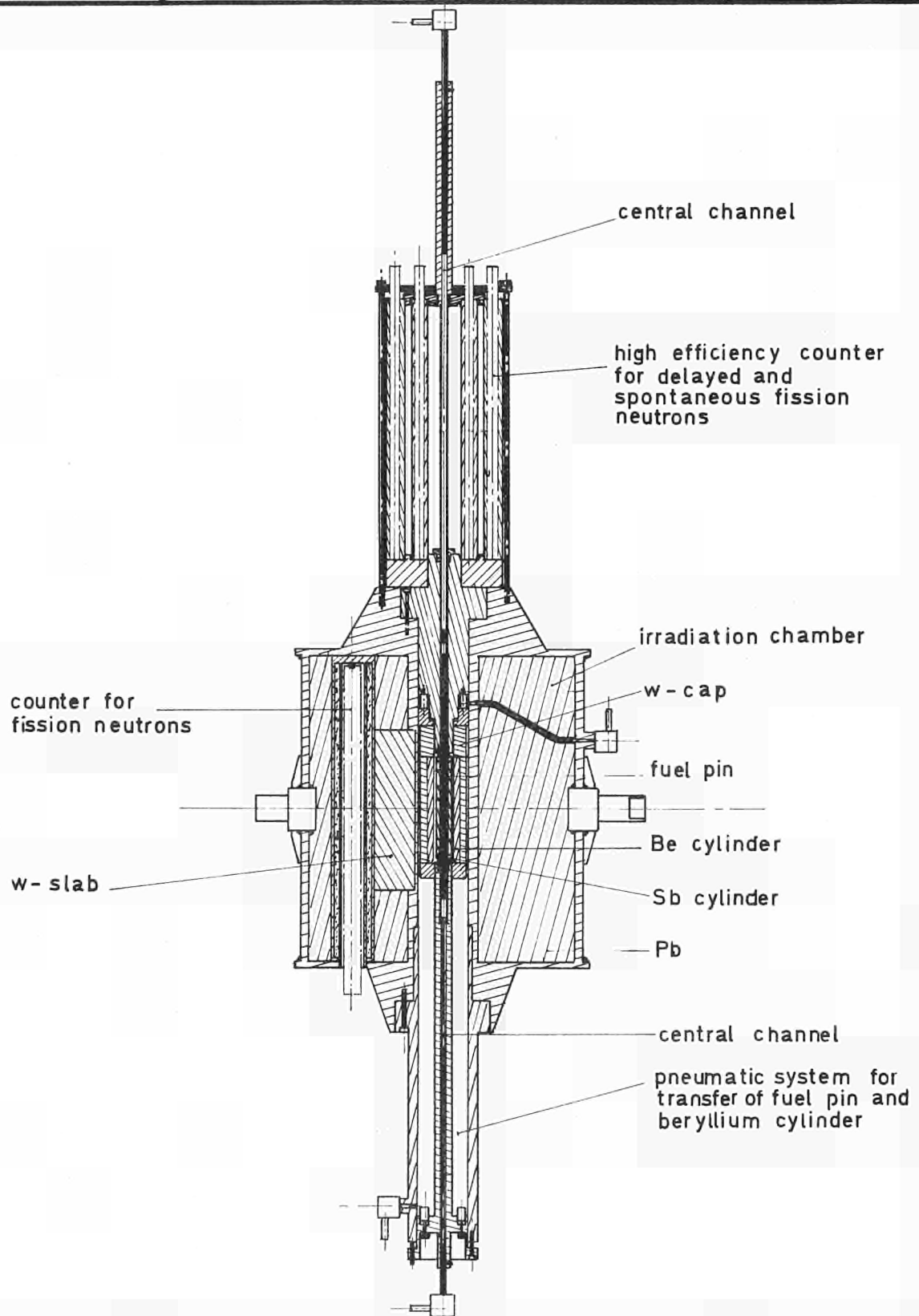




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fuel pins

Fig. 5

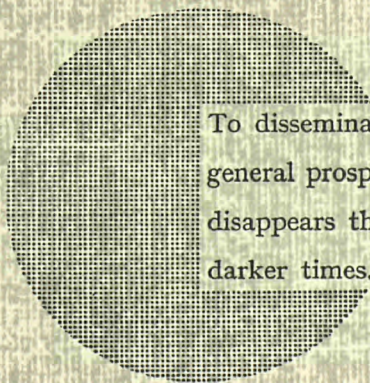


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Alfred Nobel

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