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Institut für Neutronenphysik und Reaktortechnik

Some Investigations on Non-Destructive Safeguards Techniques

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SOME INVESTIGATIONS ON NON-DESTRUCTIVE SAFEGUARDS TECHNIQUES*

K. Baumung, K. Böhnel, J. Klunker,
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Zusammenfassung

In Karlsruhe werden verschiedene zerstörungsfreie Prüfverfahren im Hinblick auf eine Entscheidung untersucht, welche Methode oder Kombination von Methoden für die Durchführung der Spaltstoffflußkontrolle am besten geeignet ist. Dabei wurden bisher diejenigen experimentell untersucht, über die im folgenden berichtet wird, nämlich Bremszeitspektrometer, Reaktivitätsmessungen, Messung der verzögerten Neutronen, aktive und passive hochauflösende γ-Spektroskopie, sowie Koinzidenzmessung von Spontanspaltungsneutronen.

Mit dem Bremszeitspektrometer wurde die Spaltrate von Brennstäben als Funktion der Energie der Neutronen im Bereich von 10 eV bis 3 000 eV gemessen. Nach Eichung gegen einen Standardstab erhält man daraus die Menge des spaltbaren Materials. Die Unterscheidung von ^{239}Pu und ^{235}U ist möglich, wenn man das Verhältnis der Spaltraten in verschiedenen Energiebereichen heranzieht. Die Genauigkeit und Reproduzierbarkeit der Methode wurden untersucht.

Reaktivitätsmessungen können mit guter Genauigkeit in kurzer Zeit durchgeführt werden, sind aber normalerweise kein betrugssicheres Verfahren zur Bestimmung des Spaltstoffgehalts. Macht man aber für jeden Stab 3 Messungen in jeweils genügend verschiedener Umgebung, kann man alle wichtigen Betrugsmöglichkeiten ausschließen, ausgenommen radiale Inhomogenitäten. Dies wurde mit einem Experiment in einer H_2O-moderierten kritischen Anordnung gezeigt.

Die Entdeckung von Spaltmaterial in einem simulierten Abfallfaß wurde mit einer gepulsten Quelle und der Messung der verzögerten Neutronen untersucht. Dabei wurde die Abhängigkeit des Ergebnisses vom Ort des Spaltmaterials im Faß und von der Dichte des Füllmaterials (Eisen und Paraffin) gemessen, wobei moderierte und nichtmoderierte Targets verwendet wurden.

Die 414 keV γ-Linie des ^{239}Pu kann für genaue passive Plutoniumbestimmungen in Brennstäben verwendet werden, sofern die Homogenität der Brennstoffverteilung in einer gesonderten Messung nachgewiesen wurde. Dies kann durch einen Intensitätsvergleich der emittierten γ-und Röntgenstrahlung geschehen. Der Gesamtspaltstoffgehalt kann mit Hilfe der während kurzer Bestrahlungen induzierten γ-Aktivität der Spaltprodukte gefunden werden. Dieses und einige ähnlichen Probleme wurden unter Verwendung eines Li-gedrifteten Germaniumdetektors studiert.

Es wurde ein hochempfindlicher Neutronendetektor gebaut, um schnell und genau die Spontanspaltung im Plutonium zu bestimmen. Dabei wurde eine Rossi-α-Koinzidenztechnik angewandt, um mit einem totzeitfreien Gerät gegen (α,n)-Prozesse zu diskriminieren.

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Abstract

Non-destructive assay techniques are studied at Karlsruhe with the purpose of identifying the optimal method or combination of methods for a practical safeguards system. The methods which have been investigated experimentally so far will be reported below:
Slowing down time spectrometer, reactivity measurements, delayed neutron measurements, active and passive high resolution γ-spectroscopy, and coincidence measurements of spontaneous fission neutrons.

The slowing time spectrometer was employed to measure the fission rate of fuel pins as a function of neutron energy in the range between 10 eV and 3,000 eV. When calibrated against a standard, this indicates the amount of fissile material. Discrimination between ^{239}Pu and ^{235}U is possible if counting rate ratios of appropriate energy ranges are taken. Accuracy and reproducibility of the method were checked.

Reactivity measurements can be made with good accuracy in a short time but are normally no tamperproof determinations of the fuel content. However, if three measurements are made for each pin in sufficiently different environments, all important methods of cheating the determination can be excluded. An exception is constituted by radial inhomogeneities. This was demonstrated in an experiment made with an H_2O-moderated critical facility.

The detection of fissile material in a simulated scrap barrel was studied with a pulsed source and the delayed neutron technique. The sensitivity of the result with respect to the location of the fissile material in the barrel and the density of the filling material(iron and paraffin) was investigated. Moderated and unmoderated targets were used.

The 414 keV γ-line of ^{239}Pu can be used for accurate passive plutonium determination in fuel pins, provided that the homogeneous distribution of fuel is checked separately.This may be done by comparing the intensities of the γ- and X-rays emitted. The total fissile material content can be found by fission product γ-counting after short-term irradiation. This and some similar problems were studied by means of a Li-drifted germanium detector.

A high sensitivity neutron counter was built to measure spontaneous fission of plutonium quickly and with high precision. A Rossi-α coincidence technique was used to discriminate against (α,n)-processes with a deadtime-free instrument.

1. INTRODUCTION

The investigations reported here were made in connection with the development of an instrumented safeguards system in which the flow of fissile material is measured at certain strategic points in the fuel cycle [1]. Non-destructive methods are required primarily at the exit of a fuel element fabrication plant to determine the amount of fissile material in newly fabricated fuel elements. Measurements on spent fuel are of secondary importance in this context, since chemical methods can be applied when the fuel is dissolved in the reprocessing plant. The methods should be tamperproof and able to discriminate between ^{235}U and plutonium.

So far, the investigations have been concentrated on the identification of the methods or combinations of methods which seem to be suited best for this purpose and measurements have been made to supplement and partly parallel the activities abroad. Instruments are considered which measure the total throughput of a fabrication plant relative to standards the composition of which is determined by destructive methods. No effort was made to develop a portable general-purpose instrument.

As a consequence of the required tamperproofness only radiation of high penetrability can be used for the measurements. Any substantial radiation absorption leads to a dependence of the results of the measurements on the spatial distribution of the fissile material in the fuel, and thus it is possible to cheat by differences in spatial distribution. This is particularly serious when complete subassemblies are subjected to measurements. Therefore, in spite of the obvious advantages of measurements on subassemblies, methods were studied also which are applicable to single fuel pins only.

Because of the large number of pins (between 10^5 and 10^6) that are measured in one inventory period the statistical accuracy of the individual pin measurement can be rather poor eventually, and 10 % will be sufficient. For subassemblies, the accuracy should be about 1 %. The time available for a pin measurement is of the order of a few minutes, whilst several hours of measurement are possible for a subassembly.

Passive and active methods were investigated and neutron and γ-ray emissions were measured. No equipment was available for γ-induced reactions.

Two sets of test pins were prepared and used in most of the measurements. They consisted of unsintered oxide pellets with a fuel density of $\rho = 6.3$ g/cm^3 and had a length of 100 cm. One set of 5.73 mm pellet diameter and with a 0.5 mm stainless steel cladding was to simulate fast reactor fuel pins (SS), the other set of 11.7 mm pellet diameter and equipped with a zircaloy cladding was to represent light water reactor pins (ST). The data of the test pins are summarized in TABLE I.

2. PASSIVE METHODS

2.1. γ-Ray Measurements

If the material to be assayed emits a characteristic radiation, the most obvious method of determining the amount of that material is to measure the intensity of this characteristic radiation.

^{239}Pu emits a γ-spectrum with a number of characteristic peaks. Unfortunately, all of them are characterized by low energy and, thus, poor penetrability. The highest energy is observed for the 414 keV γ-line which was utilized to determine the amount of ^{239}Pu in the test pins mentioned above. A 23 cm^3 Li-drifted Ge-detector was used and the test pins were positioned in front of the detector with 0.3 cm of lead in between. The results of the measurements are summarized in TABLE II; they allow to draw the following conclusions: 1) The intensity of the radiation is more than sufficient; 2) for homogeneous pins of the same density and diameter the correlation between the counting rate and the amount of ^{239}Pu is strictly linear; 3) accuracy and reproducibility are better than 0.5 % and well within the limits of statistical error.

Since no other isotope is known which emits 414 keV radiation with a long lifetime, the applicability of this radiation depends primarily on the quality of fuel homogeneity and the cladding thickness control.

For a homogeneous oxide fuel pin of 2 R = 1.0 cm pellet-diameter and ρ = 9.3 g/cm^3 about 60 % of the 414 keV γ-radiation is absorbed within the pin. However, the dependence of the counting rate on the position of fissile material in the pin is smaller than one would expect from this figure. Calculations by Weitkamp /2/ showed that for the pin of 1 cm diameter, with all plutonium concentrated close to the surface, a 20 % higher counting rate is obtained than with homogeneously distributed fuel or, in other words, with radial inhomogeneities of fuel distribution the measurement can be cheated by up to 20 % of the fissile material. For a pellet diameter of 6 mm the value is 10 % /2/. These quantities can be much reduced when the homogeneity of the material is checked separately.

In order to study the dependence of the emitted γ-spectrum on the spatial distribution of plutonium in the fuel, measurements were made with a few plutonium foils sandwiched between uranium foils to make up a total batch of 8 mm thickness. The position of the plutonium foils in the sandwich was varied and throughout the sandwich the ratio of the 414 keV peak to the 98,4 keV U $K_{\alpha 1}$-radiation and to the 94.6 keV U $K_{\alpha 2}$-radiation turned out to be extremely dependent on the plutonium foil position. This may offer a good possibility for homogeneity control.

2.2. Neutron Measurements

Due to the intensity, passive neutron counting only allows to determine ^{240}Pu by its spontaneous fission rate. The ^{239}Pu content can be deduced provided that the isotopic domposition is known. Neutrons from (α,n) reactions cannot be considered as a tamperproof signal and must be suppressed by a coincidence technique. In spite of these deficiencies, the method is attractive because of its simplicity and the high penetrability of the fission neutrons.

The coincidence counting rate is proportional to the square of the detector efficiency which, therefore, should be high. High efficiency can be achieved best with thermalized neutrons. However, in that case the relatively long neutron lifetime requires wide coincidence gates which can lead to serious dead-time losses, thus reducing the reliability of the measurement. Dead-time losses can be avoided when the delayed coincidence technique of Rossi-α-measurements is applied /3/. With adequate multi-channel analyzers every detector signal opens the coincidence gate and for the counting rate in a gate of the width Δt at delay

time t [4] this results in

$$n_{\Delta t}(t) = \gamma_1 \left[\gamma_2 \varepsilon_f^2 S_f \frac{\overline{\nu(\nu-1)}}{2} \alpha e^{-\alpha t} + (\varepsilon_f S_f + \varepsilon_s n_s)^2 \right] \Delta t$$

with

S_f = number of fissions per second

ε_f = detector efficiency

ε_s = detector efficiency for (α,n)neutrons

n_s = number of (α,n)-neutrons emitted per second

α = fundamental mode decay constant of the detector, higher modes being neglected

γ_1, γ_2 = constants which are unity for dead-time free equipment and smaller than unity in other cases.

The amplitude of the exponential term is proportional to S_f and can be used for the ^{240}Pu-determination.

Preliminary test measurements were made with the pins of TABLE I inserted in a 120 cm x 51 cm x 51 cm block of polyethylene which consisted of 3 cm x 3 cm and 1 cm x 1 cm prisms of 120 cm length. Four ^{3}He-counters were set around the pins. Measurements with different pin-to-counter distances were made and a 32 channel analyzer based on the shift register principle was used.

The amplitude of the fundamental mode decay constant was determined by a least squares fit. The results were improved by using the decay constant as an input for the fit with the value determined from data with better statistics. For comparison, an evaluation was made in which all scaler contents were summed up and the uncorrelated background was subtracted as calculated from the counting rate. This procedure corresponds to the usual method of gating but without dead-time in the trigger channel. An example of the results for a measuringtime of 20 minutes is given in TABLE III.

No substantial improvement can be seen when the amplitude of the exponential decay is utilized. However, this measurement gives more insight into instrument performance and malfunctions can be detected more easily.

3. ACTIVE METHODS

3.1. Neutron Induced γ-Ray Measurements

Counting of fission product γ-rays after short term irradiation is a standard technique of fission rate determination and can be used also for fissile material assays in fuel pins. Differences in the mass distribution of fission products from ^{239}Pu and ^{235}U indicate that the γ-ray spectra could show also characteristic differences. In order to check this, ^{235}U and ^{239}Pu samples were irradiated for 2 minutes in a thermal neutron flux of 10^6 n/cm^{2}sec and the γ-spectra for different waiting times were measured with the Ge(Li)-detector mentioned above. In general, the ^{239}Pu and ^{235}U spectra were quite similar, but for a few peaks substantial differences were observed. For illustration, two spectra are shown in Fig.1. The peak at 1248 keV is six times higher in ^{235}U than in ^{239}Pu and

the peak at 767 keV is about twice as high in ^{239}Pu as in ^{235}U, furthermore the 724 keV peak of ^{239}Pu does not appear in ^{235}U.

The counting rate in the peaks after background subtraction is about 30 counts per minute. In view of the large number of pins measured, such a low counting rate may still be acceptable (see Section 1).

The penetrability of 1 MeV radiation is much better than that of 400 keV γ-rays and the amount by which the measurement can be cheated by radial inhomogeneities is only 3 % and 6 %, respectively, instead of the 10 % and 20 % mentioned in Section 2.1. for 414 keV γ-rays.

If we can exclude serious radial inhomogeneities, a combination of passive γ-counting for the ^{239}Pu-determination and total γ-counting for determination of the sum of ^{235}U and ^{239}Pu would also be an acceptable method. With thermal neutron irradiation a simple neutron source will be of sufficient intensity.

If radial inhomogeneities are considered as a realistic possibility neutron flux depression in the pin must be avoided, which means that thermal and resonance neutron energies have to be excluded. However, in the keV-range the fission cross section is small and the neutron flux must be correspondingly higher. Here, the low efficiency of the γ-spectrometer is a serious disadvantage and one had better look for other methods, such as those discussed in the section below.

3.2. Methods Based on Fission Neutron Detection

The occurrence of neutron induced fission is a unique feature of fissile material. Thus, it is the most obvious choice to use this phenomenon for fissile material assays. Because of their high penetrability fission neutrons can be utilized best for identification. However, this requires discrimination between

(1) injected neutrons and emitted fission neutrons
(2) fission in fertile and in fissile material
(3) fission in ^{235}U and ^{239}Pu.

In addition, the requirement of transparency must be taken into account.

The characteristics which can be used for discrimination are neutron energy and time. An example of time discrimination are delayed neutron measurements. Some problems of this technique are discussed in Section 3.2.3.

An example of energy discrimination are 26 keV neutrons of an Sb-Be-source impinging upon the pin or subassembly. If fission neutrons are detected with proton recoil counters where the threshold is set at about 0.5 MeV, discrimination (1) is achieved and (2) is fulfilled automatically, only discrimination (3) requires another measurement.

The transparency of the radiation is very good. If a subassembly is rotated in front of the source - which is enclosed in lead for γ-shielding - the situation is similar to irradiation in an isotropic neutron flux. In this case, only absorption is responsible for neutron attenuation, and for a typical light water reactor subassembly the ratio of the average flux in the subassembly to the flux at the surface is $f = \frac{\bar{\phi}_{av}}{\phi_o} = 0.94$. Multiplication

brings this figure even closer to 1. With a source intensity of 10^9 n/sec ($\approx$ 200 Ci) the number of induced fissions per second is larger than the number of spontaneous fissions in plutonium with 20 % ^{240}Pu. The counting rates obtained are sufficient.

3.2.1. The Slowing Down Time Spectrometer

An alternative method is a fission rate determination with the slowing down time spectrometer /5/. Because of the time energy correlation this instrument can be considered as a neutron source with a variable spectrum. The spectrum is a broad peak with half-widths between 30 % and 100 % and the peak energy ranging from 10 keV to thermal.

Discriminations 1 and 2 are achieved as for the Sb-Be source but, in addition, ^{239}Pu and ^{235}U can be discriminated because of differences in the shape of the cross sections. In the first measurements by Seufert and Stegemann the 0.3 eV resonance of ^{239}Pu was used as a signature, but self-shielding limits its applicability to fuel with less than 1 % plutonium. Therefore, the whole range of neutron energy has been investigated now. The same equipment was used as described previously /5/. It is illustrated in Fig. 2. Fig. 3 shows the measured fission rate versus neutron energy for ^{235}U and ^{239}Pu. Four energy intervals were selected in which the counting rate ratio of plutonium to uranium is markedly different. TABLE IV shows the $^{239}Pu/^{235}U$ counting rate ratios for these intervals.

A $^{239}Pu/^{235}U$-discrimination factor of 2 is obtained and certainly can be increased through a more careful selection of intervals.

The counting rates were 100 - 1,000 counts per minute and energy interval for the pins of TABLE I. The accelerator data were 125 keV, 1 mA beam current, 4 µsec pulse length and a recurrence frequency of 1,000 cps. With cadmium covered pins for thermal neutron background suppression the recurrence frequency could be increased and with higher accelerator performance, in addition, an increase in intensity by a factor of 5 - 10 seems feasible.

A serious problem to measuring in the resonance region are self shielding effects. They were determined from the variation of the counting rates per gram with the fissile material concentration. For each neutron energy interval the inverse counting rate was plotted versus $\sqrt{M}$, where M is the amount of fissile material in the pin, and from extrapolation the value for infinite dilution was obtained. Agreement of the results with calculations was always within 10 % but mostly much better. For illustration, some data are given in TABLE V. They refer to typical characteristics of fast and thermal reactor pins: Oxide fuel of ρ = 9,3 g/cm^3 smear density with d = 5.73 mm and d = 10.0 mm pellet diameter. a is the enrichment or corresponding figure for plutonium. Use was made of the fact f depends only on ρ.a.d.

For homogeneous pins self shielding does not impose serious problems since a linear calibration curve is obtained in a counting rate versus $\sqrt{M}$ presentation. However, with radial inhomogeneities the measurement can be cheated. Fissile material close to the surface of the pin is exposed to a higher neutron flux than material in the center. Thus, when concentrated at the surface a smaller amount of material is required to give the same signal as a nominal homogeneous pin. In the resonance re-

gion the difference seems to be very small, however.

Self shielding is sufficiently small in the keV-region but with measurements only in this region it is not possible to discriminate between ^{239}Pu and ^{235}U. Discrimination can be achieved by combination with a delayed neutron measurement. Irradiation of the pins in the lead pile must be carried out separately from the pulsed measurements because of background and intensity reasons.

Measurements of the $^{238}U/^{235}U$ fission rate ratio and of the detailed spectrum between 10 keV and 200 keV showed that in the stationary spectrum of the lead pile the ^{238}U fission rate is small enough to be treated as a correction even for low enrichment fuel and that, on the other hand, the spectrum is hard enough to give good penetrability of the fission inducing neutrons [6]. Preliminary delayed neutron measurements showed that the intensity is also sufficient and overrides spontaneous fission by a factor of 5. However, measurements with the pins covered by cadmium revealed a high contribution of low energy neutrons. For ^{239}Pu 35 % of the fissions were caused by subcadmium neutrons. This is probably due to neutrons thermalized in the concrete shield of the lead pile. A better setup is required if this method is seriously considered. However, it seems to be much more convenient to determine the total fission rate with an Sb-Be source and the delayed fission rate with an accelerator source with tailored spectrum. The domain of the slowing down time spectrometer is the resonance region.

An instrument is in the final stages of industrial development by the INTERATOM company which will be used for pins of thermal reactors with low plutonium content [7]. The 0.3 eV resonance will be used for Pu/U-discrimination.

3.2.2 Reactivity Measurements

If homogeneity of the fuel pins is checked separately, thermal neutron irradiations can be applied. In this case, reactivity measurements in a critical facility would offer a possibility of fissile material determination, especially when high accuracy and short measuring time are required. An advantage is the high reliability of the facilities. One problem consists in making the measurements tamperproof.

There are various possibilities of obtaining the same reactivity signal from pins of different fissile material contents. The most important ones are:

(1) addition or subtraction of absorber
(2) exchange of ^{235}U and ^{239}Pu
(3) addition of moderator
(4) modification of the spatial distribution of the fissile material in the pin (axial or radial)

Since weight and dimensions of the pin must be correct, not much moderator substance can be added, which excludes possibility (3). (4) is excluded by the homogeneity test; so, this leaves only (1) and (2). If we perform 3 measurements for each pin in environments of different sensitivities to absorber, ^{235}U and ^{239}Pu, we can also exclude these.

A critical facility with a light water region has very low sensitivity to absorber if the absorber is put in the center of this region.

If the radius of the water region is large compared to the migration length of thermal and epithermal neutrons, the neutrons absorbed in the pin in any case stand no chance of reaching the core region again. Thus, the influence of the absorber on the reactivity is very small. However, fission neutrons which are generated in the pin are able to reach the core, provided that the dimensions of the flux trap are smaller than the slowing down length of fission neutrons. Thus, fissile material has a substantial reactivity effect.

^{235}U and Pu can be discriminated when, in addition to the measurement in a standard environment, one measurement is made with the pin surrounded by gadolinium. Gadolinium cuts off the thermal neutrons but lets the 0.3 eV neutrons penetrate. Therefore, ^{239}Pu has a much larger reactivity effect in this environment than ^{235}U.

These facts were demonstrated by measurements made with the testpins of TABLE I in a light water critical facility at GEESTHACHT /8/. The facility consists of MTR-type fuel elements and had a special element where the pins could be inserted. Three configurations were studied with different sizes of the water region and measurements were made with the pins inserted in gadolinium and cadmium covered tubes. The following results were obtained:

1. For the pins of thermal reactors (ST) reactivity varies linearly with the fissile material content, for fast reactor pins self shielding is prohibitively large.

2. In the measurements with the gadolinium tube the per gram reactivity effect of plutonium is 3.41 times higher than the value for ^{235}U. Without gadolinium this factor was 1.32. This results in a $^{239}Pu/^{235}U$-discrimination factor of 2.6.

3. The ratio of the reactivity of an absorber (nickel tube) to the reactivity of plutonium decreases rapidly with the effective radius R of the cylindrizised water zone and almost approaches zero for R = 10 cm (see Fig.4). If the absorber surrounds the fuel pin, a finite remains value. This results from a reduced fission rate in the pin due to the flux attenuation effect of the absorber. Yet, there is still a discrimination factor of 4.

4. For plutonium concentrations of 1 % or more the 0.3 eV resonance is too strongly self shielded to be useful for ^{235}U and ^{239}Pu discrimination. In this case, the reactivity determination can be combined with a delayed neutron yield measurement. Irradiation during the reactivity measurement generates enough delayed neutron precursors. The ratio of delayed neutron emission to reactivity effect is independent of self shielding and depends only on the $^{235}U/^{239}Pu$-ratio of the fuel. Measurements made with the detector described in Section 3.2.3. resulted in a $^{235}U/^{239}Pu$-discrimination factor of 1.8. However, if possible, the gadolinium tube measurement should be preferred to avoid complicated flux normalization problems.

3.2.3. Delayed Neutrons in Scrap

Delayed neutron measurements have been mentioned above several times as a possible supplementary technique for fissile material assays in fuel elements. However, the main application for which this method seems particularly well suited is the determination of fissile material in scrap. The method was pioneered by Keepin and was extensively investigated at Los Alamos /9/. Work at Karlsruhe only parallels what

has been done in the U.S. but it is hoped that it will result in some supplementary information as well.

Measurements with a simulated scrap barrel were made with a compact 14 MeV neutron generator developed by W. Eyrich /10/. Most of the measurements were made on scrap containing hydrogen. It was simulated by 5 cm x 5 cm x 10 cm prisms of paraffin which were set in a loose arrangement on circular aluminium sheets. 9 of these layers were arranged on top of each other to form a cylinder of 90 cm height and 60 cm diameter. The average paraffin density was varied between 8.8 vol.% and 26 vol.% by changing the number of paraffin prisms in the assembly. The source was located on one side of the cylinder and a semiannular array of 16 2 in. diameter BF_3 counters in paraffin was arranged on the opposite side. Small samples of ^{235}U and natural uranium were located in various positions in the "barrel" to determine the space dependence of the signal. Measurements were made also with iron prisms instead of paraffin and with an empty barrel. The influence of filters was studied also. Fig.5 shows the principal arrangement.

A comprehensive report of all measurements is being prepared /11/; therefore only some results are given here.

(a) Space Dependence:

There is a strong radial dependence of sensitivity with maximum sensitivity close to the detector. However, when two measurements are made with the barrel rotated by 180° and the average is taken, the radial distribution is flattened very much and the factor of 6 is reduced to about 20 %. For an empty or nearly empty barrel the situation is worse. In this case, one had better use a different arrangement. For instance, delayed neutron precursors could be generated in a hydrogen free environment with the barrel after irradiation placed in the center of an annular detector arrangement.

Azimuthal measurements revealed that practically the same result is obtained with continuous rotation of the barrel and with the average of two stationary barrel measurements with the barrel position differing by 180°. However, continuous rotation may cause trouble when the rotation time is comparable with the half life of delayed neutron emitters.

Axial dependence is strong but fortunately fairly insensitive to the filling of the barrel.

(b) Density Dependence

The dependence of the counting rate per gram ^{235}U on the hydrogen density also was rather marked with a sensitivity peak at about 25 vol.% paraffin for 14 MeV neutron injection. The maximum shifted towards lower densities when filters were applied.

The "add-a-gram" technique as proposed by the Los Alamos group /12/ proved to be extremely useful. The optimum position for the additional sample turned out to be immediately in front of the detector. With this position the density dependence was reduced from a factor of 7 to 10 %. Again, the situation was worse for very low hydrogen density; in that case, a different position for the sample should be chosen.

(c) Discrimination between ^{238}U and ^{235}U

To reduce fission in ^{238}U a lead filter of 20 cm thickness was placed in front of the target. This decreased the fission rate in ^{238}U so much that, for the natural uranium sample, it was lower than the fission rate from ^{235}U. Thus, if the enrichment of the material in the scrap is roughly known, a global correction for the ^{238}U-effect will be adequate.

The gravest error in the measurement will result probably from self shielding of lumped fissile material in the scrap. Even for an empty barrel 30 % of the neutron flux and 99 % of the fissions resulted from thermal and epithermal neutrons which were mostly slowed down in the detector. The situation can be improved if the irradiation is performed without the detector but it will always be a problem to get a sufficiently non-moderating environment.

With the time averaged source intensity of $Q = 10^9$ n/sec the sensitivity limit was 1 g ^{235}U. In the case of plutonium, the delayed neutron emission has to override the spontaneous fissions, which leads to a minimum average source intensity of about 10^{10} n/sec.

4. CONCLUSIONS

1. Presently, no single non-destructive method fulfilling all requirements of a safeguards instrument is available, and perhaps only a combination of methods will offer the necessary resistance to tampering.

2. For fissile material assays of fuel elements, measurements on complete subassemblies are preferred from the plant operator's point of view. When an isotropic neutron flux with neutrons of the energy $E >$ 10 keV is used for interrogation and fast neutrons are detected, the transparency seems to be adequate. A possible method is the plutonium determination from spontaneous fissions in ^{240}Pu or with the calorimeter, combined with a total fissile material determination from a fission rate measurement where fissions are induced by an Sb-Be source. A disadvantage is the fact that the isotopic composition of plutonium must be known and the plutonium determination is not really tamperproof. An alternative is a delayed neutron yield measurement where, for instance, the total fission rate is measured with the Sb-Be source and delayed neutrons with a pulsed accelerator. A Van de Graaf accelerator would be the ideal neutron source, but a 14 MeV-neutron generator with moderated target may possibly be sufficient. The difference in delayed neutron yield will then allow to determine ^{235}U and ^{239}Pu separately.

One can think of several similar techniques, for instance, with different neutron energies used for interrogation. Common to all these methods is the problem encountered by low energy neutrons backscattered from the environment.

3. If these problems turn out to be prohibitively difficult, measurements must be conducted on single pins. Several other methods are available then; for instance, those based on resonance neutron interrogation or γ-ray detection. Passive γ-counting combined with activation analysis is a simple and promising technique provided that fuel

homogeneity can be checked from low energy gamma peak ratios with sufficient accuracy. Resonance neutron counting with the slowing down time spectrometer is more complicated but needs less stringent fuel homogeneity tests.

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TABLE I Data of Test Pins

Symbol	Pu (%)	^{235}U (%)	$^{239}Pu + ^{241}Pu$ /g/	^{235}U /g/	$^{239}Pu + ^{241}Pu + ^{235}U$ /g/
ST1	0	2	0,0	11,97	11,97
ST2	0,5	2	2,78	12,00	14,78
ST3	1	2	5,53	11,89	17,42
ST4	0	2,5	0,0	15,05	15,05
ST5	0,5	2,5	2,77	14,99	17,76
ST6	0	3	0,0	18,08	18,08
SS1	10	3	14,59	4,29	18,88
SS1	10	6	14,52	8,52	23,04
SS3	10	9	14,61	12,86	27,47
SS4	10	18	14,54	25,61	40,15
SS5	12,5	0	18,20	0,99	19,19
SS6	15	0	21,76	0,96	22,72
SS7	17,5	0	25,29	0,93	26,22
SS8	25	0	36,53	0,85	37,38

Isotopic composition of Plutonium:

^{239}Pu	90.875 %
^{240}Pu	8.225 %
^{241}Pu	0.856 %
^{242}Pu	0.040 % .

TABLE II ^{239}Pu-Determination from 414 keV γ-rays

Pin	counts/ 10 min	counts/ (10 min·g ^{239}Pu)	Pin	counts/ 10 min	counts/ (10 min .g ^{239}Pu)
SS1	21617	1484	ST2	3243	1167
SS2	21201	1460	ST3	6437	1163
SS3	21665	1481	ST5	3198	1154
SS4	21432	1475			
SS5	26985	1481			
SS6	32378	1489			
SS7	37548	1483			
SS8	53939	1476			

TABLE III ^{240}Pu Determination from delayed coincidence measurements (Cd-shielded pins)

Pin	M /g/	A arbitrary units	A/M	n_c arbitrary units	n_c/M	n_t /sec^{-1}/	n_t/M
ST2	2,78	85	30.6	7.0	2.50	85	30.6
ST3	5.53	175	31.6	13,9	2.51	167	30.2
SS1	14.59	432	29.6	34.6	2.37	429	29.4
SS2	14.52	436	30.0	34.5	2.38	423	29.1
SS3	14.61	434	29.7	35.4	2.42	415	28.4
SS4	14.54	419	28.8	34.8	2.39	430	29.6
SS5	18.20	517	28.4	42.6	2.34	534	29.3
SS6	21.76	638	29.3	51.2	2.35	639	29.4
SS8	36.53	1078	29.5	85.3	2.34	1075	29.4

M = 0.9173 x mass of Pu (see Table I)

A = Amplitude of fundamental mode

n_c = Number of correlated coincidences (channel 2-20, i.e. gate from 16 to 320 /usec)

n_t = total counting rate

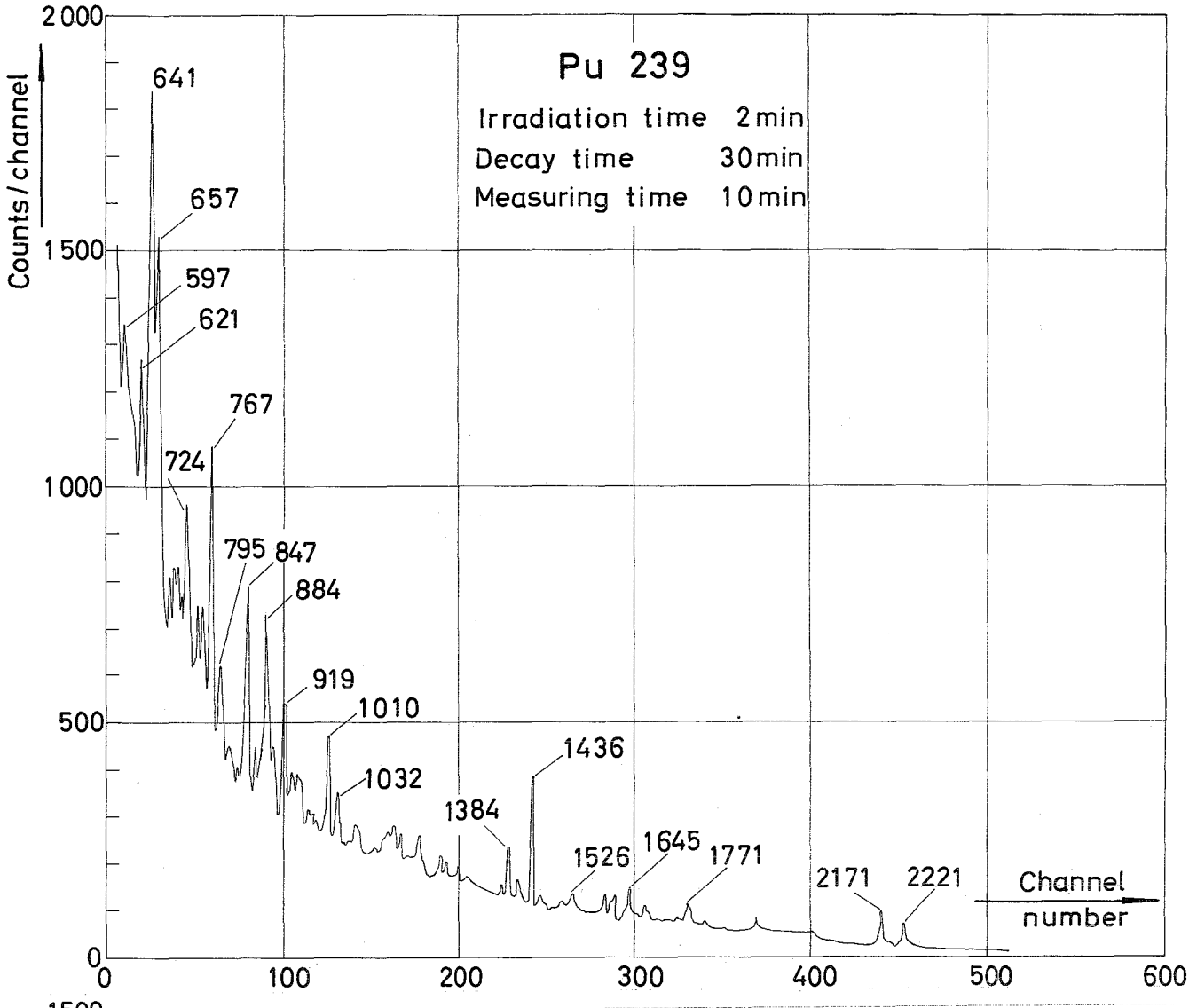
Multiplication was neglected.

TABLE IV $^{239}Pu/^{235}U$ counting rate ratios as measured in the lead pile in different neutron energy intervals

Energy range	1	2	3	4
Time interval /μsec/	7.5 - 13.5	39 - 55.5	60.5 - 78	85.5 - 131
Neutron energy /eV/	1000-3260	60 - 120	30 - 50	10.7 - 25
Pu/U counting rate ratio	0.815	1.45	0.859	1.78

TABLE V Average self shielding factors f for pins with enrichment a and pellet diameter d

Energy intervals		1	2	3	4
a(Pu)					
d = 5.73mm	d = 10.0mm				
3 %	1.73 %	0.96	0.90	0.80	0.81
4.75 %	2.73 %	0.95	0.87	0.74	0.78
9.5 %	5.45 %	0.92	0.82	0.65	0.70
14.25 %	9.15 %	0.90	0.78	0.60	0.65
a(^{5}U)					
d = 5.73mm	d = 10.0mm				
3 %	1.73 %	0.97	0.88	0.93	0.79
4.75 %	2.73 %	0.96	0.86	0.91	0.73
9.5 %	5.45 %	0.95	0.81	0.87	0.65



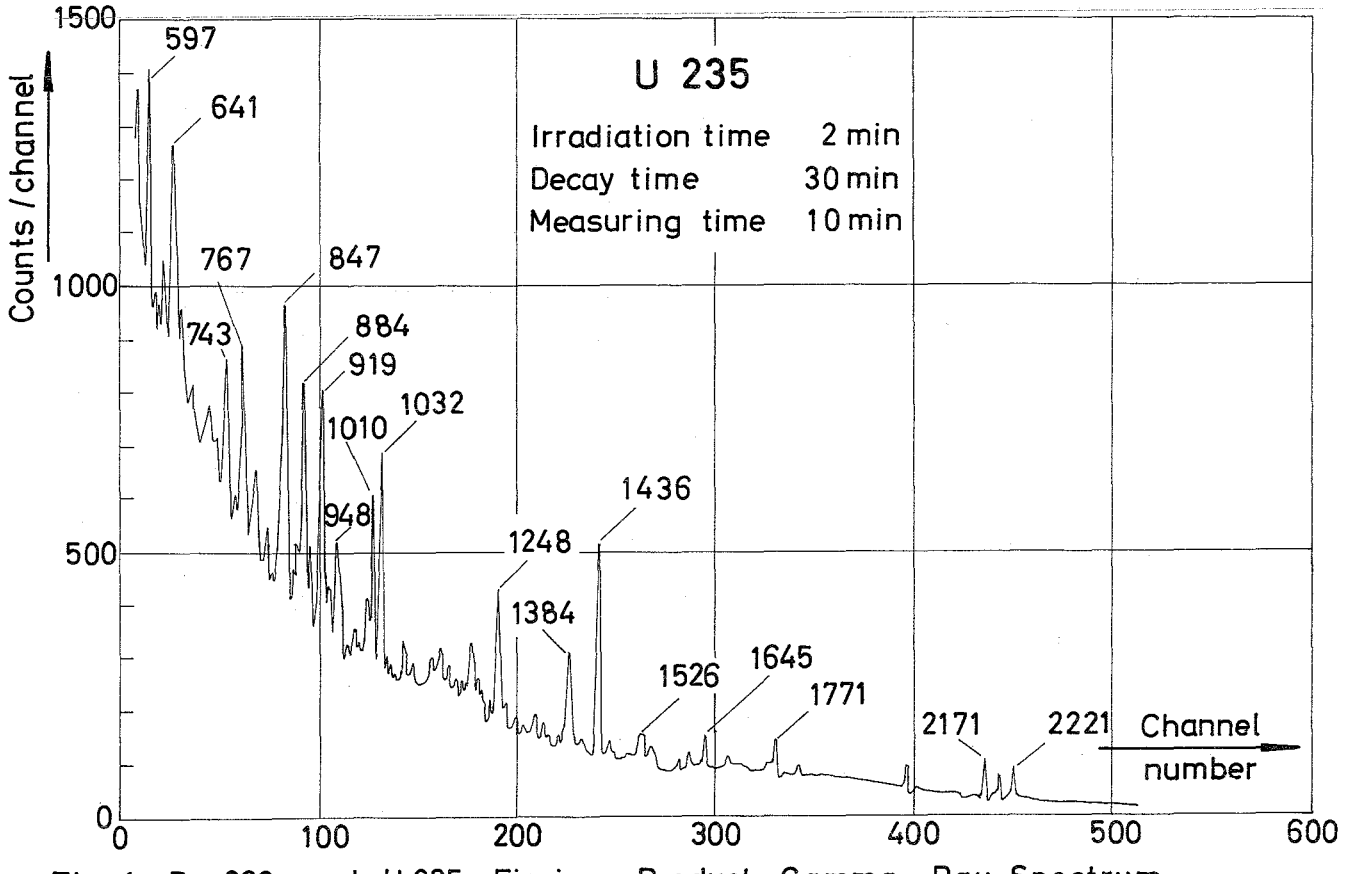


Fig. 1 Pu 239 and U 235 Fission - Product Gamma - Ray Spectrum

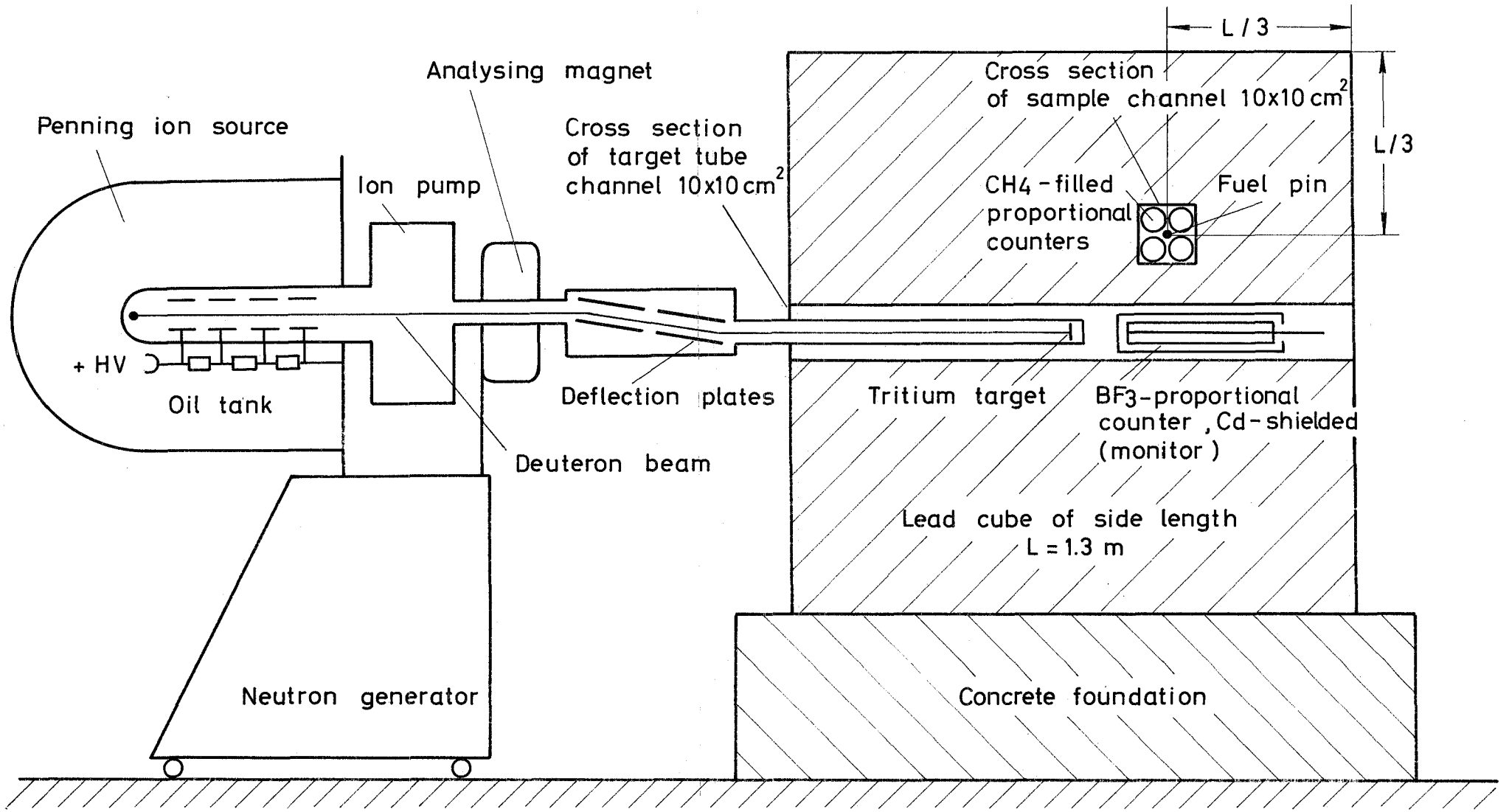
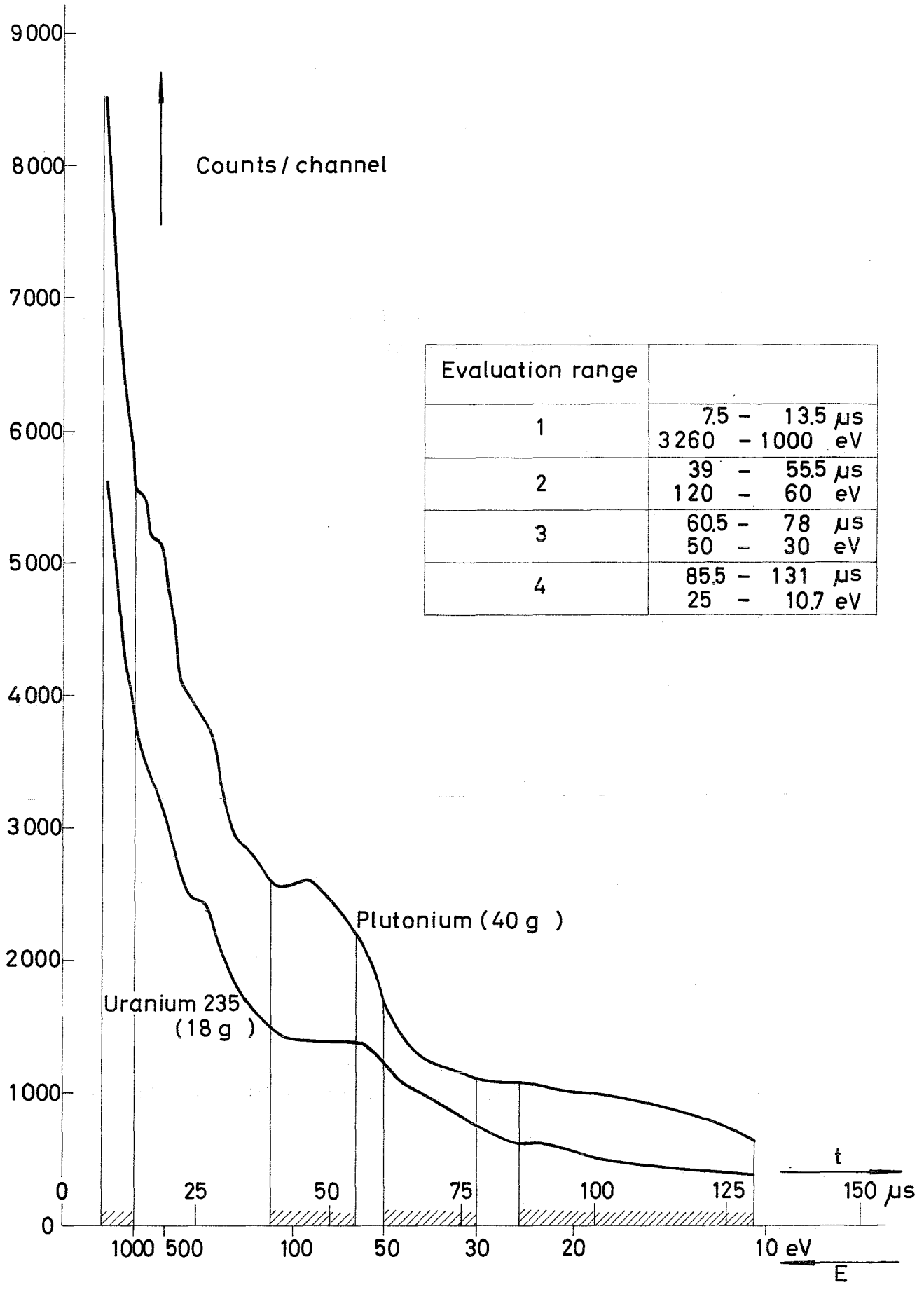


Fig. 2 Setup of Neutron Slowing Down Time Spectrometer



Evaluation range	
1	7.5 – 13.5 µs 3260 – 1000 eV
2	39 – 55.5 µs 120 – 60 eV
3	60.5 – 78 µs 50 – 30 eV
4	85.5 – 131 µs 25 – 10.7 eV

Fig. 3 Time and Energy Dependence of the Fission Rate in Fuel Pins

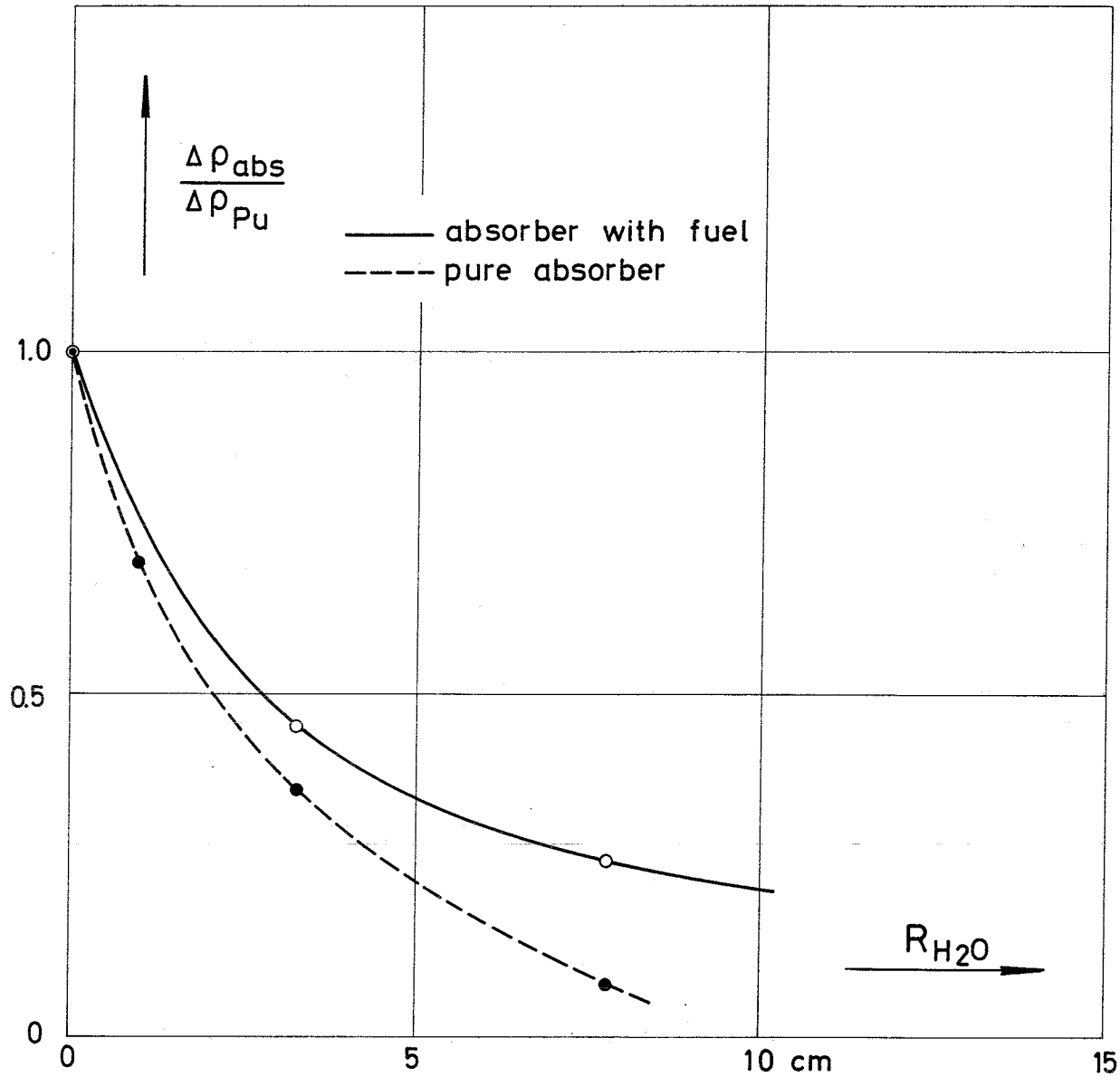
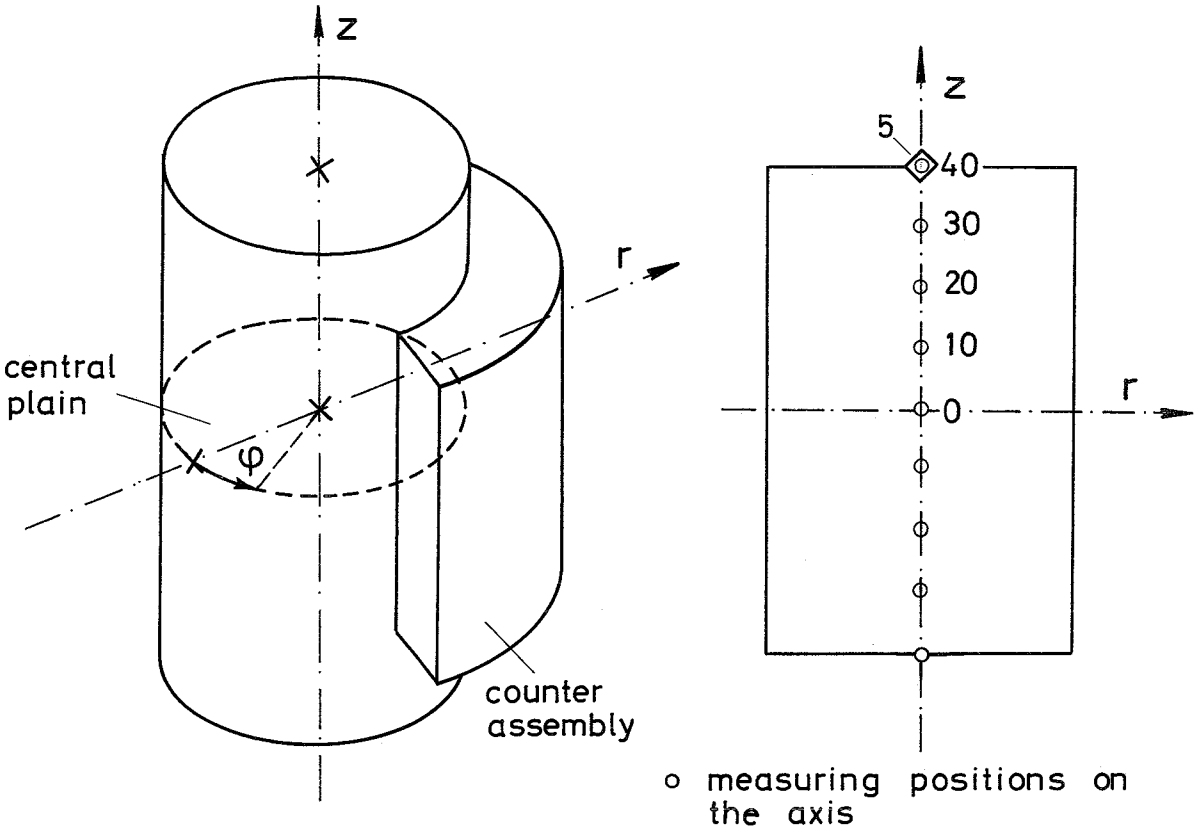


Fig. 4 Reactivity of Absorbers in Reactors with Central H_2O-Zone



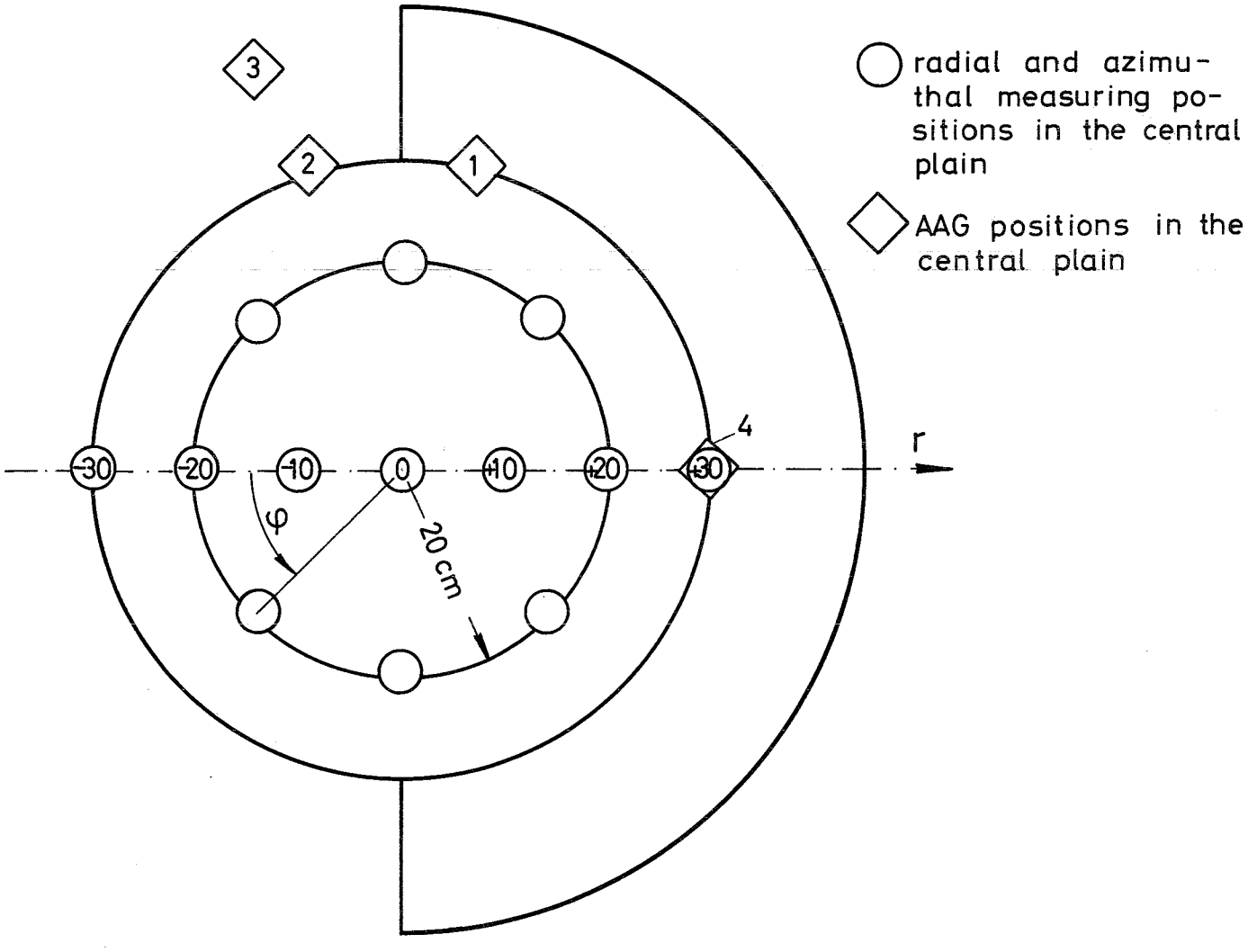


Fig. 5 Measuring Positions in Scrap Barrels