# EUR 5158 e

COMMISSION OF THE EUROPEAN COMMUNITIES

# ON THE DETERMINATION OF THE Pu 240 IN SOLID WASTE CONTAINERS BY SPONTANEOUS FISSION NEUTRON MEASUREMENTS.

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1974

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surviving fraction and accordingly $F_1^{40}$ becomes smaller. This

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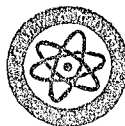
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## ABSTRACT

A non-destructive measurement system for the passive assay of plutonium in solid waste drums is described. The fission neutrons from Pu-240 spontaneous fissions are measured with a system consisting of a flexible well-type counter built up of six modular units, each containing three $^3$He-counter tubes of 50 cm length, either coupled to a variable dead-time counter containing pulse amplification, -shaping and -discrimination chains for the six neutron detection units, or to a small computer (8K memory) able to do time correlation analyses of the pulse output from the detector units. The computerized part was meant to serve mainly as a reference method for the variable dead-time counter (VDC).

The system was calibrated with PuO$_2$ standards of known isotopic composition, distributed evenly and locally in matrix materials closely corresponding to the matrix material expected in real waste drums. Calibration extended from sub-gram quantities of plutonium to several hundred grams of plutonium.

Actual waste drums were measured, of which some contained substantial amounts of fission products giving rise to high gamma doses and others unidentified $(\alpha, n)$-sources, resulting in a high neutron background. The experimental results from calibration runs are presented along with the results on the actual solid waste drums originating from the oxide production chain of the reprocessing plant. The total amount of plutonium in a drum is obtained from isotopic composition estimates.

The theoretical background and the mathematical outline of the theory is given.

the same fission event. For the described equipment $w_i$ is very

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## INTRODUCTION

Several non-destructive techniques for the determination of Pu
in solid waste based on gamma spectroscopy with a Na(I)-crystal
have been described[1, 2]. As solid waste drum assay for pluto-
nium became necessary at EUROCHEMIC, the same techniques
were applied. It soon became evident however, that this technique
was not applicable at a reprocessing plant treating a wide variety
of fuels (PWR, BWR, LWR) with a large variation in burn-up,
giving plutonium with a wide range of isotopic compositions.

The next step at EUROCHEMIC was therefore the use of a high
resolution Ge(Li)-detector coupled to a 1024-channel analyzer with
which it was possible to scan a waste drum and:

a) to "identify" the plutonium by estimating the probable isotopic
   composition,

b) to measure the content of plutonium by using selected gamma
   lines of Pu-239 and

c) to apply corrections for gamma absorption in the matrix mate-
   rial by evaluating lines of different energy from the same isotope.

Extensive description of a similar technique is published in ref. (3).

The results obtained by these measurements at EUROCHEMIC
were routinely communicated to the EURATOM Safeguard Inspec-
tion Agency in Luxembourg.

The sometimes alarmingly high quantities of plutonium reported
in these waste drums suggested to the Agency that other, indepen-
dent non-destructive techniques should be applied for control.

At this point it was recognized that the gamma-spectrometry tech-
nique only would be applicable to waste drums where the plutonium

$(y_{jk})_G$

was reasonably homogeneously distributed in material where the gamma ray attenuation would not be too high (average atomic number $\leqslant 10$).

$PuO_2$ might pass undetected by the gamma-spectrometric technique if sufficiently shielded.

Evidently, this problem is not so serious for passive neutron assay. On the contrary, this technique is much more sensitive to low atomic number materials. Till now gamma- and neutron assay are complementary techniques and from a safeguards point of view both should be applied, gamma-spectroscopy for establishing the isotopic composition of the material and neutron assay for the total quantity.

In the best interest of safeguards and reprocessing plant accountability, a cooperation was established between EURATOM, Luxembourg; EURATOM, Ispra and EUROCHEMIC with the aim to measure as many actual waste drums as possible with passive neutron assay techniques.

In Chapter 1 the description of the equipment involved is given. This chapter contains all relevant information about the performance of the detection- and the time correlation analysis systems. Emphasis is given to problems of the detector response under various conditions of samples and matrix materials and to the influence of high count rates on the coincidence rate for integral auto-correlation techniques (VDC - and other coincidence techniques). These problems are frequently ignored in practice, but this does not mean that they are negligible!

Chapter 2 deals with measurements with known samples, which serve for the determination of all parameters needed for the numerical analysis of the raw experimental data. An estimate of the diffi-

cult neutron self-multiplication effect is included. Measurements with reprocessing plant waste drums are reported in Chapter 3.

Conclusions from all reported experiments are listed in Chapter 4.

## 1. DESCRIPTION OF THE INSTRUMENTATION

A general view of the passive neutron assay equipment, which was used for measurements with 30 l waste drums is shown in Fig. 1 and a block diagram of the same is given in Fig. 2.

The various components and units of the instrumentation will be described hereafter.

### 1.1 Neutron Detector Assembly for Waste Drums

The neutron detector is built up of six modular units. These units are held together by two frames in such a way that they form a hexagonal well counter, in which a waste drum of maximum 34 cm diameter and 43 cm height can be placed. Top cover and bottom of the well are made of polyethylene plates of 6.2 cm thickness. Photographs of the assembly are shown in Fig. 3.

Each modular unit contains three $^3$He-proportional counters of 50 cm active length, 2.5 cm diameter and 6 atm gas pressure. These counters are embedded in a moderator block of polyethylene which is clad with cadmium of 1 mm thickness. Each block is covered on the internal face of the well by a steel plate of 2 cm thickness which acts as a neutron spectrum shifter and also as a gamma-shield. On the external face a 3 cm thick polyethylene plate is mounted which serves for backscattering of fast and epithermal neutrons into the counter. Both plates together provide for raising of the neutron counting efficiency and flattening of the spectral response.

where $\Delta X_{is} = p_{2i} m^2$ is the coincidence count rate due to induced

The spatial response functions $f_r(r)$ and $f_z(z)$ $[f(r, z)=f_r(r) \cdot f_z(z)]$ of the counter well for fission neutrons are shown in Fig. 4. In Fig. 5 the counter response functions $f_r(R)$ and $f_z(H/2)$, respectively averaged over radius R, and height H of cylinders are plotted.

These curves serve for the evaluation of the mean counting efficiency of cylindrical drums of various sizes.

The effect of moderating matrix material on the spatial response of the counter assembly is demonstrated in Fig. 6 for drums of various diameters and 38 cm height, filled with polyethylene grains of $0.6 \text{ g/cm}^3$ density. These curves allow for the estimation of the effect of a strongly moderating matrix on the counting efficiency. Heavy matrix material has a much smaller influence on the counter response. For example, a fission neutron source (Cf-252) has been put in the centre of a lead cylinder of 15 cm diameter and 17 cm height (ca. 30 kg) and this gave a relative increase in the counting efficiency of about 8%.

The spectral response function normalized to the Cf-252 fission neutron energy is given in Fig. 7. A neutron decay curve is shown in Fig. 8 from which the neutron die-away time of the detector assembly has been determined as $(30.0 \pm 0.3) \mu s$. There is no measurable influence of matrix material on the die-away time. Finally, in Fig. 9 the absolute fission neutron counting efficiency $\varepsilon_o(0)$ in the centre of the well is plotted as a function of discriminator settings of the VDC-unit using 1830 V detector bias voltage.

## 1.2 Variable Dead-Time Counter (VDC)

The instrument is described in ref. (4) and (5). A photograph of it is shown in Fig. 10 and the block diagram is included in Fig. 2. It consists of pulse amplification, -formation and -discrimination

and on the distribution of the fissile material within the matrix. From case to case there might be a variation of multiplication

chains for six neutron detector units, a pulse mixing circuit and one fast and four slow pulse counters with nominal dead-times $\tau_i = 16 \mu s$, $32 \mu s$, $64 \mu s$ and $128 \mu s$. The dead-time losses of the different pulse counters are analyzed in terms of count rates of neutrons from spontaneous fissioning nuclides, in this case essentially of Pu-240. The relationship between the fission neutron count rate and the measured count rates $C_i$ can be established by a simple consideration of the various components of the count rates and their balance.

Let us call
$C_o$ the count rate of the fast pulse counter and $C_o^{40}$ and $C_o^{\alpha}$ and $C_o^{bg}$ respectively the spontaneous fission neutron, $(\alpha, n)$ and background count rates of which $C_o$ is composed, we have:

$$C_o = C_o^{40} + C_o^{\alpha} + C_o^{bg}$$

With analogous definitions we write the equations for the count rates $C_i$ of the pulse counters with the dead-time $\tau_i$ as:

$$C_i = C_i^{40} + C_i^{\alpha} + C_i^{bg}$$

In order to determine $C_o^{40}$ from $C_o$ and $C_i$, we consider the following balance of count rates. From $C_o^{40}$ counts per unit time a part $(C_i^{40})$ will trigger the counter (i) with dead time $\tau_i$. Because the fission neutrons are emitted in groups (at the average 2.2 per event), there will be a fraction $F_i^{40}$ of $C_i^{40}$ correlated with the trigger pulses $C_i^{40}$ which we call the "true coincidence count rate".

Moreover, there will be a loss of $C_i \tau_i C_o^{40}$ counts due to the dead-time $\tau_i$ of the counter (i), which we call "accidental coincidence count rate". Analogous relationships are obtained for $C_o^{\alpha}$, $C_o^{bg}$ and $C_i^{\alpha}$, $C_i^{bg}$ respectively, but $F_i^{\alpha}$ and $F_i^{bg}$ are zero, unless neutron multiplication is taken into account. Neglecting the latter for the moment

we have:

$$C_o^{40} = C_i^{40}(1 + F_i^{40}) + C_i \tau_i C_o^{40} \qquad (1.a)$$

$$C_o^\alpha = C_i^\alpha \qquad\qquad + C_i \tau_i C_o^\alpha \qquad (1.b)$$

$$C_o^{bg} = C_i^{bg} \qquad\qquad + C_i \tau_i C_o^{bg} \qquad (1.c)$$

$$C_o = C_i + C_i^{40} F_i^{40} + C_i \tau_i C_o \qquad (2)$$

From eq. (1.a) and (2) we obtain

$$C_o^{40} = \left( C_o - \frac{C_i}{1 - C_i \tau_i} \right) \cdot \frac{1 + F_i^{40}}{F_i^{40}} \qquad (3)$$

$$C_o^{40} = K_i X_i \qquad (3.a)$$

with

$$K_i = \frac{1 + F_i^{40}}{F_i^{40}} \qquad (4)$$

$$X_i = C_o - \frac{C_i}{1 - C_i \tau_i} = C_o + \frac{1}{\tau_i - \frac{1}{C_i}} \qquad (5)$$

$K_i$ can be determined by a calibration measurement with a small Pu-240 sample (metallic plutonium) where $C_o^{40}$ and $X_i$ can be measured directly. It can also be calculated from nuclear parameters (fission neutron emission multiplicity) and instrumental parameters (counting efficiency and die-away time) as shown in ref. (6). The effective dead-time $\tau_i$ is equal to the nominal dead-time ($\tau_{i_o}$) minus the discriminator paralysis time $\vartheta$, averaged over the six counting chains. The averaged $\vartheta$ is obtained from the eq. (6).

$$\sum_{k=1}^{6} \frac{C_{o_k}}{1 - C_{o_k} \vartheta_k} = \frac{C_o}{1 - C_o \vartheta} \qquad (6)$$

where $C_{o_k}$ means the pulse rate of chain k,

$\vartheta_k$ the paralysis time of chain k, and

$C_o = \sum\limits_{k=1}^{6} C_{o_k}$ the total pulse rate. Thus $\vartheta$ becomes:

$$\vartheta = \frac{\sum\limits_{k=1}^{6} \dfrac{C_{o_k}}{1-C_{o_k}\vartheta_k} - C_o}{C_o \sum\limits_{k=1}^{6} \dfrac{C_{o_k}}{1-C_{o_k}\vartheta_k}} \qquad (7)$$

For symmetric distribution of the pulse rate, i.e. $C_{o_k} = \dfrac{C_o}{6}$ and $\vartheta_k \equiv Const \equiv \vartheta_o$, we obtain $\vartheta = 1/6\,\vartheta_o$.

$\vartheta_k$ depends on the pulse shape and the discrimination level, as shown in Fig. 11. In Fig. 12 are plotted the effective $\tau_i$-values applicable for symmetric distribution of the pulses ($C_o = \dfrac{C_o^i}{6}$, $\vartheta_k = const = \vartheta_o$) as a function of the discriminator settings. These values are derived from measurements with a pure $(\alpha, n)$ source. Eq. (5) gives for this case

$$(X_i = 0)\tau_i = \frac{1}{C_i} - \frac{1}{C_o} \qquad (8)$$

For asymmetric distributions of the pulses, formula (7) can be used for corrections.

$$\tau_i' = \tau_i + \Delta\vartheta$$

$$\Delta\vartheta = (\tfrac{1}{6}\,\vartheta_o - \vartheta)$$

Unfortunately, $F_j^{40}$ and therefore $K_i$, depend on the count rate $C_i$ itself. This is due to the fact that the higher the count rate, the more probable is the case that a spontaneous fission event occurs during the closure of the counter (i). Then a fraction of the emitted neutron population dies away before the counter may be retriggered by the

surviving fraction and accordingly $F_i^{40}$ becomes smaller. This
effect is taken into account by a linear correction term given be-
low

$$K_i(C_i) = K_{io}(1 + b_i C_i) \tag{9}$$

It is more convenient to include this correction term into the $X_i$

$$X_{io} = X_i(1 + b_i C_i) \tag{10}$$

and to write

$$C_o^{40} = K_{io} \cdot X_{io} \tag{11}$$

where $K_{io}$ is now a constant.

The dependence of $b_i$ on the neutron die-away time $1$ of the detec-
tor is shown in Fig. 13. These curves have been obtained from Monte
Carlo simulation of the VDC by means of the computerized system
for time correlation analysis, which is described in the following sec-
tion.

The experimental determination of the $b_i$-values is explained in
section 2.2 in connection with the calibration of the instrument.

## 1.3 "Computerized System" for Time Correlation Analysis of Fission Neutrons

The computerized system is described in Ref. (6). This system is
generally useful for the (a) simulation and (b) measurement of the
time distribution of pulses from detector-sample arrangements, and
analysis of the time correlations among these pulses. In the present
investigations it serves mainly as a reference method for passive
neutron assay by the VDC-instrument.

curves. In the plot of the variation of the detector response as

The "simulation" is performed by the Monte Carlo-method. Input data are:

i) source parameters: amount, half-life, probability distribution function of neutron emission multiplicity ($p_v$-distribution) of spontaneous neutron emitters (spontaneous fission or $(\alpha, n)$-reactions),

ii) multiplication parameters: probabilities for induced fission reactions in fissile isotopes of the sample and the respective neutron emission multiplicities for the various sources,

iii) neutron detector parameters: detection probabilities and die-away time of neutrons from the various sources.

Output data are:

i) Time distribution of neutron detection pulses,

ii) Time correlation analysis as Rossi-$\alpha$-method, VDC-method and other well known pulse correlation methods.

A schematic flow diagram is shown in Fig. 14. In the case of a "measurement", the time distribution of neutron detection pulses is produced by a converter, which converts the detector pulses in their real arrival time. 8 Inputs for detector pulses are provided and each of them is labelled $(1, 2, \ldots, 8)$. The data (time) are stored firstly in an 8-stage buffer memory from which they are transferred into the 8K-memory of the mini-computer (LABEN 70). The buffer memory acts as a "de-randomizer" of the time-distribution of pulses and allows for reduction of signal losses during the transfer cycles into the computer memory. A block diagram of this instrumentation is given in Fig. 15.

As a reference method for passive neutron assay techniques, we choose the well known Rossi-$\alpha$-method, which does not require any correction. The Rossi-$\alpha$ technique allows for the separation of correlated counts from uncorrelated counts out of the time-distribution

In reality there will always be a superposition of both the self-multiplication and the matrix effects, which sometimes may add

of neutron detection pulses. This is done by summing up all pulses within a time interval, $\Delta t$, which follows each individual detector pulse and subtraction of the uncorrelated counts within $\Delta t$, which are computed from the total count rate $C_o$ as $C_o \Delta t$. In the picture of a coincidence counter, and this is also true for the VDC, this would mean that any detector pulse may open a coincidence gate or close all pulse counters (i) respectively. In this way there would be no need for correction either for true coincidence losses, during the time the coincidence gate was opened or the input of pulse counter was closed respectively, or for the variation of the $F_i$ (correlated counts per pulse due to fission neutrons) with count rate $C_i$ (linear correction factor $b_i$). This feature of the Rossi-$\alpha$ technique is therefore a possibility for testing the correction formula of the VDC method. The second weak point of the VDC is the variation of the dead times $(\tau_i)$ due to unsymmetric repartition of the detector pulses among the 6 pulse counting chains. Also this uncertainty is eliminated in the Rossi-$\alpha$ analysis, just by introducing a blank of 4 $\mu$sec at the beginning of the time interval $\Delta t$ to be analyzed.

## 2. MEASUREMENTS WITH STANDARD SAMPLES IN 30 L. DRUMS

The measurements with known $(PuO_2)$ samples served for the determinations of

i) $F_\infty^{40}$ = average number of neutron detector pulses correlated with the first neutron detector pulse from a Pu-240 spontaneous fission event (integral autocorrelation).

ii) $b_i$ = linear correction factors,

iii) calibration curves for the VDC instrument and the computerized system.

The standard samples covered the range of Pu-240 equivalent mass $(m_{eq}^{40})$ between 0.09 g and 54 g.
$m_{eq}^{40}$ is defined according to eq. (3) as

$$m_{eq}^{40} = m^{40} \sum_k \frac{F_i^k}{F_i^{40}} \frac{1+F_i^{40}}{1+F_i^k} \frac{m^k}{m^{40}} \frac{n_o^k}{n_o^{40}} \qquad (12)$$

(k includes 40)

upper index 40, k stays for Pu-240 or isotope named "k" respectively.

$m^{40, k}$ = mass of spontaneous fission isotope Pu-240 or "k" respectively.

$n_o^{40, k}$ = number of neutrons emitted per g of Pu-240 or "k" respectively.

$F_i^{40, k}$ = correlated counts per trigger of counter with dead-time $\tau_i$ by a Pu-240 neutron or "k" neutron respectively.

If $F_i^k \simeq F_i^{40}$ that means: all isotopes have nearly the same fission neutron multiplicities, we can simplify eq. (12) as

$$m_{eq}^{40} \simeq m^{40} \sum_k \frac{m^k}{m^{40}} \frac{n_o^k}{n_o^{40}} \qquad (12.a)$$

This condition is nearly fulfilled for all isotopes of our interest, i.e. Pu-238, Pu-242 and U-238.

The Pu-standard samples were put into normal 30 l drums and mixed up with typical matrix materials, like gloves, cleaning papers, etc.

## 2.1 Determination of $F_\infty^{40}$

$F_\infty^{40}$ is defined by the equations (13) and (14)

$$F_i^{40} = F_\infty^{40} w_i (1-e^{-\tau_i/1}) = F_\infty^{40} \cdot \alpha_i (i = 1, 2, 3, 4) \qquad w_i \leqslant 1$$
$$(13)$$

$w_i$ corrects for the effect of having more than one trigger $(C_i^{40})$ from

(6)    BIRKHOFF, G., BONDAR, L.; "Computerized System for

the same fission event. For the described equipment $w_i$ is very close to 1;

$$C_o^{40} = X_{io} \frac{F_\infty^{40} \alpha_i + 1}{F_\infty^{40} \alpha_i} = X_{ko} \frac{F_\infty^{40} \alpha_k + 1}{F_\infty^{40} \alpha_k} \quad (k \neq i) \quad (14)$$

from which we obtain

$$F_\infty^{40} = \frac{\dfrac{X_{ko}}{\alpha_k} - \dfrac{X_{io}}{\alpha_i}}{X_{io} - X_{ko}} = \frac{1}{\alpha_k} \frac{1 - \dfrac{y_{ik}}{A_{ik}}}{y_{ik} - 1} \quad (15)$$

with $y_{ik} = \dfrac{io}{X_{ko}}$ and $A_{ik} = \dfrac{\alpha_i}{\alpha_k}$

The $y_{ik}$ are the ratios of dead-time corrected true coincidence count rates $X_{io}/X_{ko}$, according to eq. (10).

The $\alpha_i$ (or $A_{ik}$) can be calculated from the known $\tau_i$ and 1 or they can be determined directly from measurements of the $F_i^{40}$ with a small Pu-metal sample (low count rate, no $(\alpha, n)$ reactions) from the following relationships, derived from eq. (3):

$$F_i^{40} = \frac{C_o^{40}(1 - C_i^{40} \tau_i)}{C_i^{40}} - 1 \quad (16)$$

$$\frac{\alpha_i}{\alpha_k} = \frac{F_i^{40}}{F_k^{40}} \quad (17)$$

for $\tau_k / 1 \gg 1$ we have $\alpha_k \approx 1$, $F_k^{40} \approx F^{40}$ and $\alpha_i \approx \dfrac{F_i^{40}}{F_\infty^{40}}$.

The parameter $F_\infty^{40}$ itself is defined by the following formula:

$$F_\infty^{40} \approx \varepsilon \frac{1}{2} \left\{ \left( \frac{\displaystyle\sum_{\nu=0}^{N} \nu^2 p_\nu^{40}}{\displaystyle\sum_{\nu=0}^{N} \nu p_\nu} \right) - 1 \right\} \quad (18)$$

where $p_\nu$ is the probability of emitting $\nu$ neutrons in a fission event of $Pu^{40}$.

In principle the measurement of $F_\infty^{40}$ allows the matrix effect, i. e. the influence of the matrix material on the detection efficiency $\varepsilon$, to which $F_\infty^{40}$ is directly proportional, to be estimated. However, the $F_\infty^{40}$ determined by the $y_{ik}$ and $A_{ik}$ measurements according to formula (15) is very sensitive to the uncertainties of those quantities $\delta y_{ik}$, $\delta A_{ik}$ respectively.

$$\delta F_\infty^{40} = \frac{1}{\alpha_k} \left\{ \frac{\frac{1}{A_{ik}} - 1}{(1 - y_{ik})^2} \ \delta y_{ik} + \frac{y_{ik}}{A_{ik}^2 (1 - y_{ik})} \ \delta A_{ik} \right\} \qquad (19)$$

The behaviour of this function in the range $1 \geqslant \varepsilon \geqslant 0.03$ is shown in Fig. 16.

Generally, the $A_{ik}$ can be determined to about $\delta A_{ik} = \pm 0.01$. The uncertainty of the $y_{ik}$ may also be about $\delta y_{ik} = \pm 0.01$ for not too high count rates ($C_o$). Under such conditions the relative uncertainty of the actual $F_\infty^{40} = 0.066$ (for $\varepsilon = 0.062$) amounts to about

$$\frac{\delta F_\infty^{40}}{F_\infty^{40}} = \pm 17\%.$$

The precision of the $F_\infty^{40}$-measurement can be improved significantly, by raising the detector efficiency $\varepsilon$. In Table I the measured $\alpha_i$, $A_i$, $y_i$ and $F_\infty^{40}$ values are summarized.

## 2.2 Determination of the Linear Correction Factors $b_i$

The linear correction factor $b_i$ takes into account the variation of the $F_i^{40}$ with the count rate $C_i$ as defined by eq. (9) and eq. (10), $b_i$ is determined by the relationship:

$$\frac{(y_{ik})_{C_i}}{(y_{ik})_o} = \frac{1+b_i C_i}{1+b_k C_k} \tag{20}$$

$$b_i = \frac{1}{C_i}\left\{\frac{(y_{ik})_{C_i}}{(y_{ik})_o}(1+b_k C_k) - 1\right\} \tag{21}$$

where $(y_{ik})_{C_i}$ means $\frac{X_i}{X_k}$ for $C_i$

and $(y_{ik})_o$ means $\frac{X_i}{X_k}$ extrapolated for $C_i = 0$. (see Fig. 17)

For $k = 1$, we have $b_k C_k < b_i C_i \ll 1$ and $b_k$ does not need to be known very exactly, for the determination of the $b_i$ ($i = 2, 3, 4$). In fact $b_k$ was determined directly from $X_k$-measurements with a small Pu-metal sample as a function of the count rate $C_k$, which was varied by adding $(\alpha, n)$ source-neutrons. These experimental results agreed very well with a Monte Carlo-calculation by means of the computerized system.

The experimental $b_i$-values are derived from the plots $X_i/X_1$ as functions of $C_i$ of Fig. 17.

A summary of the experimental and Monte Carlo-results is given in Table 2.

## 2.3 Calibration Curves

Calibration curves for VDC- and Rossi- -techniques are given in Fig. 18.

Least squares fits to polynomials showed that the best fitting functions are of the type

$$m_{eq}^{40} = a_{oi} + a_{1i}X_i + a_{2i}X_i^2 \text{ for VDC technique and}$$

$$m_{eq}^{40} = a_{o\alpha} + a_{1\alpha}X_\alpha + a_{2\alpha}X_\alpha^2 \text{ for Rossi-}\alpha\text{-technique.}$$

All curves from VDC-techniques are essentially of the same shape. This is more directly demonstrated by the best-fitting parameters, for which the following equations must hold:

$$a_{oi} = a_{ok} \qquad (22)$$

$$a_{2k} = a_{2i}\left(\frac{a_{1k}}{a_{1i}}\right)^2$$

Equivalence between VDC and Rossi-$\alpha$ -techniques is checked by analogous conditions:

$$a_{oi} = a_{o\alpha} \qquad (23)$$

$$a_{2\alpha} = a_{2i}\left(\frac{a_{1\alpha}}{a_{1i}}\right)^2$$

The experimental results confirm the conditions (22) fairly well but not as well for the conditions (23). This indicates a certain systematic discrepancy between VDC and Rossi-$\alpha$ analysis. In principle, the Rossi-$\alpha$-technique should be more exact than VDC owing to the uncertainties of the linear correction factor $(b_i)$ and the effective dead-time $(\tau_i)$ of the VDC technique. These uncertainties cause systematic errors for high count rates, i.e. large Pu-240 amounts, which are affecting the coefficients $a_{2i}$ of the quadratic correction terms of the calibration curves. These terms take account of various non-linear phenomena which are mainly due to the neutron multiplication and slowing down properties of the standard samples. The slowing down properties are essentially determined only by the hydrogenous matrix materials. As the matrix was always the same for all standard samples, we may conclude that the quadratic correction term of the calibration curve mainly describes the neutron multiplication effect.

## 2.3.1 Neutron Multiplication Effects

Neutron multiplication by induced fission processes, in Pu-239 and Pu-241 mainly, changes the neutron emission multiplicity. Both the spontaneous fission and the $(\alpha, n)$ neutrons are contributing to this effect. An exact treatment of this problem is very difficult and it is unresolvable for unknown samples. Nevertheless, a working hypothesis for estimating the magnitude of this effect can be established. In the case of $PuO_2$ standard samples, the ratio of spontaneous fission to $(\alpha, n)$ neutron emission and therefore $X_{is}/C_{os} \equiv r_i$ is nearly constant. The production of induced fission neutrons,

$$P_s = (\overline{\Phi} \cdot \nu \Sigma_f \cdot V)_s$$

should be proportional to the square of the Pu-mass, because both the mean neutron flux $\overline{\Phi}$ and the mean induced fission cross section $\Sigma_f V$ are proportional to the Pu-mass (V means the volume of the sample).

In the case of Pu-waste, the assumption about the constant spontaneous fission to $(\alpha, n)$ neutron emission ratio cannot be maintained. but we may account for this by a correction factor on the neutron flux. If we assume that the neutron flux $\overline{\Phi}$ is proportional to the count rate $C_o$, this flux correction factor is equal to the ratio of the count rate $C_o$ of the unknown sample to the count rate $C_{os}$ of the standard sample. The production of induced fission neutrons in the unknown sample can be written therefore as

$$P = P_s \cdot \frac{C_o}{C_{os}} \simeq km^2 \frac{C_o}{C_{os}} = km^2 r_i \frac{C_o}{X_i} \tag{24}$$

If we further assume that the coincidence count rate, $\Delta X_i$, due to induced fission events, is proportional to P, we obtain

$$\Delta X_i = \Delta X_{is} \frac{C_o}{C_{os}} \simeq p_{2i} \cdot m^2 \frac{C_o}{C_{os}} = p_{2i} m^2 r_i \frac{C_o}{X_i} \tag{25}$$

where $\Delta X_{is} = p_{2i} m^2$ is the coincidence count rate due to induced fissions in the standard sample.

The relationship between $p_{1i}$ and the best fitting parameters of the calibration curves $a_{1i}$ and $a_{2i}$ is obtained from the equations ($a_{oi} \approx 0$, $p_{oi} \approx 0$ neglected)

$$m = a_{1i} X_i + a_{2i} X_i^2 \tag{26}$$

$$X_i = p_{1i} m + p_{2i} m^2 \tag{27}$$

or

$$m = \frac{1}{2} \frac{p_{1i}}{p_{2i}} \left( \sqrt{1 + \frac{4 p_{2i}}{p_{1i}^2} X_i} - 1 \right) \tag{28}$$

For $\dfrac{4 p_{2i}}{p_{1i}^2} X_i = 4 p_{2i} \cdot a_{1i}^2 X_i \ll 1$ $(p_{1i} = \dfrac{1}{a_{1i}})$

the root expression of equation (28) can be expanded in a Taylor series ignoring the third order term. Comparison of coefficients gives

$$p_{2i} \approx - \frac{a_{2i}}{a_{1i}^3}$$

In all assumptions mentioned above, the correction for the neutron multiplication effect of unknown samples, i.e. the difference relative to standard samples is equal to

$$\Delta m_{eq}^{40} = a_{2i} X_i^2 (1 - \frac{C_o}{C_{os}}) = a_{2i} X_i^2 (1 - r_i \frac{C_o}{X_i}) \tag{29}$$

The various parameters for the calculation of $\Delta X_i$ and $\Delta m_{eq}^{40}$ are summarized in Table 3.

In reality, the neutron multiplication effect is much more complicated. It depends strongly on the moderating properties of the matrix

0.19 0.01 0.12 0.05 0.13 0.72 0.04 0.46 0.11 0.01 0.02 0.10 0.09 0.13 0.09 0.05 0.21 0.06 0.05 0.24 0.04 0.04 0.004

and on the distribution of the fissile material within the matrix.
From case to case there might be a variation of multiplication
effect by order of magnitude.

### 2. 3. 2 Matrix Effects

The influence of a polyethylene matrix which is one of the strong-
est moderating media, on the counting efficiency of the detector as-
sembly, is shown in Fig. 6, especially by the curve $\varepsilon_o(R)/\varepsilon_o(0) \overline{f. (R)}$.

From this curve one can estimate the maximum possible variation
of the counting efficiency for a given net weight of the waste drum,
just assuming that all the matrix material in the waste is composed
of highly moderating materials. If a variation of the counting effi-
ciency within $\pm 10\%$ (corresponding to $\pm 20\%$ variation of the calibra-
tion curves) is considered as tolerable, the net weight of the waste
drum should not exceed 3 kg. On the other hand, weakly moderating
matrix does not perturb so much the counting efficiency. A multiple
of the 3 kg limit can be allowed for high Z materials as matrix, like
Fe, Cu, Pb, etc. without exceeding the $\pm 10\%$ limit for the variation
of the counting efficiency. Therefore, it is highly desirable to mea-
sure the counting efficiency directly. A possibility of such a mea-
surement is explained in the section 2. 1. Owing to the dependence
of the precision of this method on the counting efficiency itself, as
shown in Fig. 16, this method is restricted to high efficiency detec-
tors. Moreover, owing to the increase of systematic errors of the
coincidence count ratios $(X_i/X_k)$ with count rate $C_o$, this method is
also limited to low count rates of about smaller than $5. 10^3$ cps.
These limitations of the method are anyhow adapted to the problem,
because waste materials contain generally small amounts of Pu.

### 3. MEASUREMENTS WITH UNKNOWN QUANTITIES OF Pu IN 30 1 WASTE DRUMS

After calibration of the instruments, 134 measurements with waste

drums have been performed during a period of about 3 months.
No calibrations of the instruments were done during this period.
As far as the VDC instrument is concerned, no breakdown has been
observed. On the other hand, an electronic defect of the "fast data
acquisition unit" (de-randomizer) was discovered after 1 month
operating time. This defect caused systematic errors in the ana-
lysis and it was detected because of discrepancies between the VDC
and Rossi-$\alpha$(computer) results. For this reason the "de-randomizer"
had to be repaired and the 18 most interesting waste drums were re-
measured. These measurements finally serve for comparison be-
tween the Rossi-$\alpha$ method (reference method) and the VDC method.
It is clear that this comparison gives a direct answer about the va-
lidity of the VDC analysis method.

## 3.1 Results

The results of the analysis of all 134 measurements are summa-
rized in Tables 4 and 5.

In Table 4 the results of 18 measurements for comparison between
VDC and Rossi-$\alpha$ analysis are given. Table 5 represents the VDC
results for all measurements which have been made. The data are
presented in the same sequence as they have been measured over a
3 months' period. Some of the waste drums have been measured se-
veral times, for example drum code E 281-2 has been measured 4
times during 3 months and no drift of the results is observed.
The quoted error limits are the sum of three sources of error:

i) counting statistics $\Delta_1 X_i$

ii) uncertainty and variation of the effective counter dead times $\Delta_2 X_i$

iii) uncertainty of the linear correction factor $b_i$.

$\Delta_1 X_i$ is calculated from the standard error of the count rate $C_o$,
which is equal to $\sqrt{C_o}$ and from the statistical error of the dead
times losses $(C_o - C_i)$ which is equal to $\sqrt{C_o - C_i}$. With these standard

errors we obtain:

$$\Delta_1 X_i = \pm \left\{ \frac{1+C_i b_i}{\sqrt{t}} \left( \sqrt{C_o} - \frac{\sqrt{C_o} - \sqrt{C_o - C_i}}{1 - C_i \tau_i} \right) \right\} \tag{30}$$

where t means the total counting time,

$\Delta_2 X_i$ and $\Delta_3 X_i$ are obtained from error propagation of $\Delta \tau_i$ and $\Delta b_i$ respectively.

$$\Delta_2 X_i = \pm \left( \frac{C_i}{1 - C_i \tau_i} \right) \left( 1 + C_i b_i \right) \Delta \tau_i \tag{31}$$

$$\Delta_3 X_i = \pm \frac{C_i}{1 + C_i b_i} X_i \Delta b_i \tag{32}$$

$$\Delta X_i = \pm \left( |\Delta_1 X_i| + |\Delta_2 X_i| + |\Delta_3 X_i| \right) \tag{33}$$

From $\Delta X_i$ we calculate the uncertainty of the Pu eq. mass as

$$\Delta m = \pm \left( a_{1i} \Delta X_i + 2a_{2i} X_i \Delta X_i \right) \tag{34}$$

where $a_{1i}$ and $a_{2i}$ are the parameters of a best fitting parabola to the calibration curves.

No corrections have been applied for neutron multiplication- and matrix effects. The methods for estimating these effects, (as explained in sections 2. 3. 1 and 2. 3. 2) are not sufficiently accurate.

## 3. 2 Discussion of the Results

The important question concerning the validity of the VDC concept for time correlation analysis has been rigorously tested by the comparison with a standard method, the Rossi-$\alpha$ technique. The time correlation analysis of the waste measurements also is an extremely hard testing in so far as the neutron emission rate of some of the waste drums is very high and in addition the ratio of correlated to uncorrelated counts is very low. For instance the waste drum E 250 gave a count rate of $2.2 \times 10^4$ cps and the ratio of correlated

to uncorrelated counts was about 80% smaller as compared to the $PuO_2$ standard samples.

The results of the comparison between the VDC and Rossi-$\alpha$ analysis, as given in Table 3, show that both methods are fully equivalent, because there is a complete agreement of all results within the error limits. Generally, the error limits of the VDC analysis are decreasing with increasing dead-time $\tau_i$. There is also a certain systematic "drifting" of the values for the Pu equivalent mass which is growing up with $\tau_i$. This effect is due to the systematic variation of the effective dead-time $\tau_i$, for unsymmetric repartition of the count rate $C_o$ among the six pulses counting chains, as explained in section 1.2. This variation $\Delta\vartheta$ is always negative and tends to underestimate the coincidence count rate $X_i$ by

$$\Delta X_i = - C_i C_o \Delta \vartheta.$$

This error is generally increasing with the count rate $C_o$ and relatively decreasing with increasing $\tau$.

The second important point is the reproducibility of the results and also this point has been checked rigorously for the VDC. The results of VDC measurements in Table 4 demonstrate that there is full agreement within the quoted error limits for all measurements during a 3 months' operation without recalibration of the instruments without rejecting any data of measurements which were routinely made and processed.

The third point of main interest concerning the matrix effect, can not be considered as resolved completely. From all investigations there is however, a general statement possible about this problem. This allows waste drums to be classified by their measured net weight into two categories. One for which the maximum systematic error limits can be assigned and another category where the error limits might be exceeded. This simply means that a maximum possible error can be deduced from the net weight of the drum, assuming that all the matrix material is composed of highly neutron moderating material like polyethylene, which makes the maximum perturbation of the calibration

curves. In the plot of the variation of the detector response as
a function of quantity of polyethylene as matrix material in Fig. 6
we see an oscillation of this function from positive to zero and fi-
nally to negative perturbations, with increasing amount of matrix
material. This behaviour is a characteristic of the detection as-
sembly. It is quite reasonable to set the tolerance level of the ma-
trix effect equal to the maximum positive perturbation, which is
about 10% and to set the maximum net weight limit at the negative
perturbation of -10%, which is equal to about 3 kg. The maximum
bias can be calculated from the actual distribution of the net weight
between 0 and 3 kg just by taking a weighted average over the per-
turbation functions in Fig. 6. Moreover, it must be kept in mind, that
a variation of the detection probability (response) by $\pm 10\%$ results
in about $\pm 20\%$ perturbation of the coincidence count rate, i.e. of
the calibration factor.

The next step would be to determine the matrix perturbation of
the waste drums which exceed the 3 kg limit. For this purpose we
consider the determination of $F_{\infty}$ according to eq. (15). Here we
need a precision for $F_{\infty}$ at least within $\pm 10\%$. This could be ob-
tained by a detection head with at least 0.2 counting efficiency as
shown in section 2.1 and Fig. 16. This possibility will be investi-
gated in detail. The present detection head with 0.062 counting effi-
ciency is not suitable for the required precision of $F_{\infty}$.

A difficult point is the neutron multiplication effect; from what
has been explained in section 2.3.1, it might be important for waste
drums with large Pu content and high neutron flux. An extreme exam-
ple for this problem is the drum E 250 (50 g Pu- 240 - equivalent and
$2.2 \times 10^4$ cps).

The correction for self-multiplication according to eq. (29) amounts
to -35% for VDC results and -20% for Rossi-$\alpha$ results. These figures
are however, very rough estimates.

In reality there will always be a superposition of both the self-multiplication and the matrix effects, which sometimes may add up or compensate each other. No clear statement is possible from the experimental data.

## 4. CONCLUSIONS

The reported measurements of the Pu-240 content in waste drums from the EUROCHEMIC reprocessing plant represent a severe test on the usefulness of passive neutron assay techniques. The analysis of the experimental data allows for the definitions of the capabilities and limitations of the instruments involved in this exercise.

1. The VDC method is equivalent to the Rossi-$\alpha$ method for all investigated waste 'rums, corresponding to a maximum neutron count rate of $2.2 \times 10^4$ cps.

2. The VDC instrument has proved the highest degree of stability and operational reliability.

3. The matrix effect can be estimated from the net weight of the waste drums. Detailed investigations are required for the evaluation of the corrections from experimental data. The procedure is explained in section 2.3.2 of this paper.

4. The neutron multiplication effect is a severe source of error for drums with large amount of Pu and high neutron flux.

5. The detection limit of the VDC method is about 1 mg Pu-240 equivalent for a 15´ measuring time and low background of epithermal and fast neutrons.

6. For a maximum of 3 kg of any kind of matrix material and a maximum of about $10^4$ cps the accuracy of the measured Pu-240 equivalent mass should stay within $\pm 20\%$ relative error limits.

For the drums measured in this work, EUROCHEMIC had
a good estimate of their isotopic composition, both from admi-
nistrative control (drum origin, time of filling the drum, mass
spectrometric data on the material at the exit points) and from
high resolution gamma-spectrometric measurements on drums
and standards (see ref. (3)).

Thus, the measurements performed in this work can be used
with confidence for accountability and safeguards purposes, since
the total plutonium content is obtained by multiplying the found
Pu-240 content with the appropriate factor.

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## 6. TABLES

TABLE 1 - Experimental Data for Evaluation of $F_{\infty}^{*}$ (see eq. (15) from Measurements with $PuO_2$-samples.

| i | $\alpha_i$ | $A_{i1}$ | $y_{i1} = \dfrac{a_{11}}{a_{1i}}$ | $F_{\infty}$ |
|---|---|---|---|---|
| 1 | $0.373 \pm 0.002$ | 1.000 | 1.000 | - |
| 2 | $0.615 \pm 0.002$ | $1.649 \pm 0.010$ | $1.614 \pm 0.01$ | $0.088 \pm 0.050$ |
| 3 | $0.862 \pm 0.003$ | $2.311 \pm 0.015$ | $2.214 \pm 0.01$ | $0.093 \pm 0.030$ |
| 4 | $0.981 \pm 0.004$ | $2.630 \pm 0.018$ | $2.523 \pm 0.01$ | $0.072 \pm 0.018$ |

$*$ from measurements with Pu-metal standard $F_{\infty} = 0.066 \pm 0.002$

TABLE 2 - Calculated and Experimental Linear Correction Factors $(b_i)$

| i | $b_i$ ($\mu sec$) | |
|---|---|---|
| | calculated value | experimental value |
| 1 | $8.0 \pm 1.6$ | $8.0 \pm 3$ |
| 2 | $19.0 \pm 1.7$ | $16.0 \pm 3$ |
| 3 | $32.0 \pm 2.0$ | $28.0 \pm 3$ |
| 4 | $42.0 \pm 2.0$ | $38.0 \pm 3$ |

TABLE 3 - Parameters for Estimation of the Neutron Multiplication Effect According to eq. (25), (29).

| i | $a_{1i}$ [g sec] | $a_{2i}$ [g sec$^2$] | $P_{2i}$ [g$^{-2}$ sec$^{-1}$] | $r_i$ |
|---|---|---|---|---|
| 1 | 0.6056 | - 0.00088 | 0.00396 | 0.0155 |
| 2 | 0.3752 | - 0.00034 | 0.00647 | 0.0251 |
| 3 | 0.2736 | - 0.00018 | 0.0088 | 0.0342 |
| 4 | 0.2400 | - 0.00013 | 0.0094 | 0.0387 |
| Rossi-$\alpha$ | 0.3324 | - 0.00015 | 0.00408 | 0.0255 |

TABLE 4 - COMPARISON OF VDC- AND ROSSI-α-TECHNIQUES

| Run No. | Sample Code | $C_0$(cps) | Pu-240 equivalent mass (g) | | | | Rossi-α Results |
| | | | VDC-Results for | | | | |
| | | | $\tilde{\tau}_1$ | $\tilde{\tau}_2$ | $\tilde{\tau}_3$ | $\tilde{\tau}_4$ | |
|---|---|---|---|---|---|---|---|
| 1 | E 250 | 22,100.5 | 45.6 ± 18.5 | 50.4 ± 12.8 | 50.8 ± 9.8 | 53.8 ± 3.3 | 51.1 ± 5.9 |
| 2 | E 272-2 | 22,401.8 | 45.2 ± 19.0 | 51.9 ± 13.2 | 52.4 ± 10. | 45.9 ± 8.4 | 56.5 ± 4.1 |
| 3 | E 243-3 | 12,944.2 | 16.8 ± 6.3 | 18.9 ± 4.3 | 20.3 ± 3.4 | 21.6 ± 3.0 | 18.0 ± 2.2 |
| 4 | E 281-2 | 8,720.3 | 33.9 ± 3.5 | 34.3 ± 2.5 | 34.9 ± 2.0 | 36.8 ± 1.8 | 36.9 ± 1.2 |
| 5 | E 244-3 | 1,959.4 | 9.01 ± 0.34 | 9.11 ± 0.26 | 9.14 ± 0.23 | 9.02 ± 0.23 | 9.19 ± 0.23 |
| 6 | E 255-2 | 2,626.4 | 6.69 ± 0.46 | 6.86 ± 0.35 | 6.92 ± 0.30 | 6.85 ± 0.30 | 6.68 ± 0.32 |
| 7 | E 245 | 4,877.9 | 16.3 ± 1.26 | 16.7 ± 0.93 | 16.7 ± 0.72 | 17.0 ± 0.71 | 17.0 ± 0.64 |
| 8 | E 283-2 | 3,143.0 | 7.10 ± 0.60 | 6.71 ± 0.44 | 7.32 ± 0.38 | 7.21 ± 0.37 | 7.07 ± 0.39 |
| 9 | E 137 | 949.0 | 4.89 ± 0.15 | 4.96 ± 0.12 | 4.81 ± 0.10 | 4.90 ± 0.11 | 4.87 ± 0.11 |
| 10 | E 243-2 | 3,425.9 | 5.70 ± 0.66 | 6.20 ± 0.49 | 6.25 ± 0.41 | 5.49 ± 0.40 | 5.93 ± 0.43 |
| 11 | E 277-2 | 4,374.6 | 8.94 ± 0.99 | 9.19 ± 0.72 | 9.25 ± 0.60 | 9.36 ± 0.57 | 9.95 ± 0.57 |
| 12 | E 183-2 | 6,670.2 | 28.2 ± 2.2 | 27.9 ± 1.6 | 29.3 ± 1.3 | 27.9 ± 1.2 | 30.1 ± 0.90 |
| 13 | E 271-2 | 1,399.3 | 8.13 ± 0.23 | 8.16 ± 0.18 | 8.40 ± 0.16 | 8.59 ± 0.17 | 8.72 ± 0.17 |
| 14 | E 208-1 | 1,819.1 | 9.47 ± 0.318 | 9.70 ± 0.25 | 9.90 ± 0.22 | 9.98 ± 0.22 | 9.81 ± 0.22 |
| 15 | E 269-1 | 4,923.0 | 29.0 ± 1.44 | 29.9 ± 1.01 | 29.8 ± 0.91 | 29.6 ± 0.82 | 30.3 ± 0.62 |
| 16 | E 174 | 1,437.3 | 6.29 ± 0.23 | 6.48 ± 0.18 | 6.46 ± 0.16 | 6.72 ± 0.16 | 6.76 ± 0.17 |
| 17 | E 301 | 5,442.8 | 46.9 ± 1.9 | 45.9 ± 1.5 | 45.77 ± 1.2 | 45.5 ± 1.0 | 45.8 ± 0.67 |
| 18 | E 305 | 2,702.4 | 13.7 ± 0.55 | 13.1 ± 0.42 | 13.3 ± 0.36 | 13.4 ± 0.35 | 13.6 ± 0.32 |
| | $\sum\limits_{i=1}^{18} m$ | | 340.8 g 93% | 356.8 g 97% | 361.8 g 98% | 359.6 g 98% | 368.3 g 100% |
| | $\sum\limits_{i=3}^{18} m$ | | 250.0 g 96% | 254.5 g 98% | 258.6 g 99% | 259.9 g 100% | 260.6 g 100% |

TABLE 5 - VDC-Results

| Run No. | Sample Code | $C_o$(cps) | Pu-240 Equivalent (g) | | | |
|---|---|---|---|---|---|---|
| | | | $\tau_1$ | $\tau_2$ | $\tau_3$ | $\tau_4$ |
| 1 | E-151 | 3484.98 | 24.98 ± 0.76 | 25.13 ± 0.58 | 25.92 ± 0.49 | 26.16 ± 0.46 |
| 2 | E-243-2 | 3398.1 | 6.69 ± 0.62 | 6.82 ± 0.45 | 6.69 ± 0.37 | 6.84 ± 0.36 |
| 3 | E-246 | 2465.5 | 3.96 ± 0.37 | 4.06 ± 0.27 | 3.87 ± 0.23 | 4.24 ± 0.23 |
| 4 | E-247-2 | 815.7 | 2.71 ± 0.09 | 2.65 ± 0.07 | 2.63 ± 0.06 | 2.60 ± 0.07 |
| 5 | E-250 | 21920.0 | 35.96 ± 16.4 | 43.52 ± 10.6 | 43.26 ± 8.1 | 44.26 ± 6.91 |
| 6 | E-272-1 | 472.5 | 2.91 ± 0.06 | 2.88 ± 0.05 | 2.87 ± 0.04 | 2.82 ± 0.04 |
| 7 | E-272-3 | 202.4 | 1.38 ± 0.03 | 1.37 ± 0.02 | 1.35 ± 0.02 | 1.32 ± 0.02 |
| 8 | E-275-1 | 469.8 | 2.69 ± 0.06 | 2.77 ± 0.04 | 2.75 ± 0.04 | 2.65 ± 0.04 |
| 9 | E-275-2 | 14438.2 | 18.06 ± 7.8 | 21.49 ± 5.3 | 20.31 ± 4.1 | 22.11 ± 3.5 |
| 10 | E-277-2 | 4363.0 | 9.69 ± 0.93 | 9.95 ± 0.67 | 10.18 ± 0.55 | 10.13 ± 0.51 |
| 11 | E-281-1 | 15031.9 | 47.79 ± 7.64 | 50.83 ± 5.2 | 51.1 ± 3.9 | 50.35 ± 3.4 |
| 12 | E-281-2 | 8979.9 | 34.59 ± 3.21 | 36.60 ± 2.3 | 36.1 ± 1.8 | 36.27 ± 1.5 |
| 13 | E-283 | 12813.6 | 29.9 ± 6.2 | 33.7 ± 4.2 | 32.7 ± 3.3 | 30.82 ± 2.9 |
| 14 | E-283-3 | 5136.5 | 15.5 ± 1.3 | 15.6 ± 0.91 | 15.8 ± 0.73 | 15.7 ± 0.67 |
| 15 | E-284-1 | 2135.5 | 8.62 ± 0.33 | 8.85 ± 0.25 | 9.10 ± 0.22 | 9.13 ± 0.20 |
| 16 | E-284-2 | 860.7 | 3.15 ± 0.10 | 3.17 ± 0.08 | 3.27 ± 0.08 | 3.27 ± 0.07 |
| 17 | E-151 | 3513.5 | 24.9 ± 0.7 | 25.4 ± 0.58 | 26.0 ± 0.49 | 26.3 ± 0.45 |

Table 5 - contd.

| | | | | | | |
|---|---|---|---|---|---|---|
| 18 | E-243-2 | 3409.7 | 6.29 ± 0.62 | 6.63 ± 0.45 | 6.71 ± 0.37 | 6.76 ± 0.36 |
| 19 | E-246 | 2465.1 | 3.89 ± 0.37 | 3.94 ± 0.27 | 3.96 ± 0.23 | 4.41 ± 0.23 |
| 20 | E-247-2 | 824.5 | 2.56 ± 0.09 | 2.59 ± 0.075 | 2.60 ± 0.07 | 2.56 ± 0.07 |
| 21 | E-250 | 21677.7 | 46.2 ± 15.2 | 48.0 ± 10.2 | 46.6 ± 7.81 | 50.5 ± 6.54 |
| 22 | E-272-1 | 474.6 | 2.83 ± 0.06 | 2.84 ± 0.05 | 2.82 ± 0.04 | 2.83 ± 0.04 |
| 23 | E-272-3 | 202.3 | 1.37 ± 0.03 | 1.34 ± 0.02 | 1.32 ± 0.02 | 1.32 ± 0.02 |
| 24 | E-275-1 | 472.6 | 2.84 ± 0.06 | 2.72 ± 0.04 | 2.73 ± 0.04 | 2.70 ± 0.04 |
| 25 | E-275-2 | 14438.3 | 15.0 ± 7.3 | 18.9 ± 5.3 | 21.8 ± 4.1 | 17.9 ± 3.6 |
| 26 | E-277-2 | 4389.8 | 10.3 ± 0.95 | 10.2 ± 0.68 | 10.3 ± 0.56 | 10.4 ± 0.52 |
| 27 | E-281-1 | 15061.8 | 52.9 ± 7.4 | 53.2 ± 5.2 | 53.0 ± 3.9 | 52.6 ± 3.4 |
| 28 | E-281-2 | 9041.3 | 34.6 ± 3.2 | 35.4 ± 2.3 | 35.6 ± 1.8 | 35.8 ± 1.6 |
| 29 | E-283 | 12866.0 | 29.5 ± 6.2 | 32.3 ± 4.3 | 32.6 ± 3.3 | 32.6 ± 2.9 |
| 30 | E-284-1 | 2121.3 | 8.51 ± 0.32 | 8.51 ± 0.25 | 8.51 ± 0.21 | 8.77 ± 0.02 |
| 31 | E-284-2 | 871.4 | 3.32 ± 0.10 | 3.23 ± 0.08 | 3.26 ± 0.07 | 3.31 ± 0.07 |
| 32 | E-283-3 | 5191.5 | 15.2 ± 1. | 15.9 ± 0.92 | 16.1 ± 0.75 | 16.2 ± 0.68 |
| 33 | E-274-1 | 574.4 | 2.66 ± 0.07 | 2.60 ± 0.05 | 2.60 ± 0.05 | 2.69 ± 0.68 |
| 34 | E-130 | 394.3 | 2.33 ± 0.05 | 2.21 ± 0.04 | 2.20 ± 0.03 | 2.22 ± 0.03 |
| 35 | E-255-2 | 2557.8 | 7.38 ± 0.41 | 7.25 ± 0.30 | 7.22 ± 0.26 | 7.15 ± 0.25 |
| 36 | E-274-2 | 7628.1 | 13.1 ± 2.4 | 13.8 ± 1.7 | 13.9 ± 1.3 | 14.1 ± 1.2 |
| 37 | E-277* | 2257.3 | 5.41 ± 0.33 | 5.49 ± 0.25 | 5.40 ± 0.21 | 5.33 ± 0.21 |
| 38 | E-269-1 | 4378.2 | 29.6 ± 1.24 | 30.0 ± 0.92 | 30.4 ± 0.76 | 30.4 ± 0.68 |
| 39 | E-199-1 | 322.1 | 1.98 ± 0.04 | 1.96 ± 0.03 | 1.97 ± 0.03 | 1.96 ± 0.03 |
| 40 | E-277-1 | 848.8 | 1.82 ± 0.09 | 1.82 ± 0.07 | 1.82 ± 0.06 | 1.82 ± 0.07 |

Table 5 - contd.

| | | | | | | |
|---|---|---|---|---|---|---|
| 41 | E-279 | 700.2 | $2.19 \pm 0.07$ | $2.15 \pm 0.06$ | $2.13 \pm 0.05$ | $2.05 \pm 0.05$ |
| 42 | E-208-2 | 897.9 | $3.64 \pm 0.10$ | $3.67 \pm 0.08$ | $3.71 \pm 0.07$ | $3.64 \pm 0.08$ |
| 43 | E-282 | 758.9 | $1.42 \pm 0.08$ | $1.41 \pm 0.06$ | $1.40 \pm 0.05$ | $1.45 \pm 0.06$ |
| 44 | E-220 | 585.5 | $3.50 \pm 0.07$ | $3.44 \pm 0.06$ | $3.48 \pm 0.05$ | $3.51 \pm 0.05$ |
| 45 | E-252 | 8239.4 | $19.6 \pm 2.8$ | $19.7 \pm 1.9$ | $20.4 \pm 1.5$ | $19.8 \pm 1.3$ |
| 46 | E-244-2 | 3359.6 | $11.5 \pm 0.64$ | $11.5 \pm 0.47$ | $11.6 \pm 0.39$ | $11.6 \pm 0.37$ |
| 47 | E-177 | 491.8 | $3.16 \pm 0.06$ | $3.14 \pm 0.05$ | $3.10 \pm 0.04$ | $3.06 \pm 0.04$ |
| 48 | E-209 | 265.8 | $1.30 \pm 0.03$ | $1.28 \pm 0.02$ | $1.30 \pm 0.02$ | $1.31 \pm 0.02$ |
| 49 | E-178 | 265.7 | $1.41 \pm 0.03$ | $1.35 \pm 0.02$ | $1.35 \pm 0.02$ | $1.34 \pm 0.02$ |
| 50 | E-283-2 | 3163.5 | $6.98 \pm 0.56$ | $7.22 \pm 0.41$ | $7.55 \pm 0.34$ | $7.29 \pm 0.33$ |
| 51 | E-226 | 1045.9 | $5.88 \pm 0.13$ | $5.88 \pm 0.10$ | $5.80 \pm 0.09$ | $5.76 \pm 0.09$ |
| 52 | E-245 | 4809.9 | $17.7 \pm 1.2$ | $13.1 \pm 0.84$ | $18.3 \pm 0.68$ | $18.6 \pm 0.62$ |
| 53 | E-272-2 | 21863.9 | $50.5 \pm 15.0$ | $52.7 \pm 10$ | $52.7 \pm 7.61$ | $54.0 \pm 6.5$ |
| 54 | E-137 | 942.7 | $5.23 \pm 0.12$ | $5.35 \pm 0.09$ | $5.26 \pm 0.08$ | $5.31 \pm 0.08$ |
| 55 | E-156 | 921.9 | $5.22 \pm 0.12$ | $5.31 \pm 0.09$ | $5.32 \pm 0.08$ | $5.19 \pm 0.08$ |
| 56 | E-282[*] | 218.5 | $1.21 \pm 0.03$ | $1.18 \pm 0.02$ | $1.16 \pm 0.02$ | $1.18 \pm 0.02$ |
| 57 | E-183-2 | 6609.3 | $29.6 \pm 1.96$ | $30.0 \pm 1.41$ | $29.6 \pm 1.13$ | $29.8 \pm 1.00$ |
| 58 | E-275 | 275.8 | $1.62 \pm 0.04$ | $1.61 \pm 0.03$ | $1.61 \pm 0.02$ | $1.58 \pm 0.02$ |
| 59 | E-174 | 1436.5 | $6.72 \pm 0.19$ | $6.69 \pm 0.15$ | $6.78 \pm 0.13$ | $6.73 \pm 0.01$ |
| 60 | E-243-3 | 12868.5 | $15.9 \pm 6.35$ | $17.2 \pm 4.3$ | $19.7 \pm 3.3$ | $19.3 \pm 2.9$ |
| 61 | E-248 | 2777.1 | $6.07 \pm 0.45$ | $6.56 \pm 0.34$ | $6.46 \pm 0.28$ | $6.71 \pm 0.28$ |
| 62 | E-202 | 1122.3 | $1.88 \pm 0.12$ | $1.82 \pm 0.09$ | $1.82 \pm 0.08$ | $1.83 \pm 0.09$ |
| 63 | E-285-1 | 3286.9 | $7.70 \pm 0.59$ | $8.10 \pm 0.43$ | $7.97 \pm 0.36$ | $8.11 \pm 0.35$ |

Table 5 - contd.

| 64 | E-244-3 | 1927.4 | 5.67 ± 0.27 | 7.93 ± 0.22 | 9.64 ± 0.20 | 9.42 ± 0.19 |
|----|---------|--------|-------------|-------------|-------------|-------------|
| 65 | E-131-3 | 138.9 | 0.45 ± 0.02 | 0.45 ± 0.01 | 0.45 ± 0.01 | 0.42 ± 0.01 |
| 66 | E-244-1 | 1368.04 | 5.25 ± 0.18 | 5.45 ± 0.14 | 5.41 ± 0.12 | 5.49 ± 0.12 |
| 67 | E-146-1 | 515.7 | 3.23 ± 0.06 | 3.15 ± 0.05 | 3.13 ± 0.05 | 3.17 ± 0.05 |
| 68 | E-208-1 | 1810.4 | 10.1 ± 0.27 | 10.3 ± 0.21 | 10.4 ± 0.19 | 10.6 ± 0.18 |
| 69 | E-278 | 5588.4 | 8.66 ± 1.39 | 8.83 ± 0.97 | 9.03 ± 0.78 | 8.72 ± 0.72 |
| 70 | E-215 | 530.3 | 2.07 ± 0.06 | 2.03 ± 0.04 | 2.02 ± 0.04 | 2.04 ± 0.04 |
| 71 | E-191-2 | 3593.3 | 24.8 ± 0.79 | 24.5 ± 0.60 | 24.3 ± 0.50 | 24.0 ± 0.46 |
| 72 | E-270 | 1318.2 | 3.76 ± 0.16 | 3.52 ± 0.12 | 3.60 ± 0.11 | 3.55 ± 0.11 |
| 73 | E-152-2* | 134.3 | 0.157 ± 0.014 | 0.162 ± 0.011 | 0.100 ± 0.01 | 0.170 ± 0.01 |
| 74 | E-276 | 238.6 | 1.39 ± 0.03 | 1.38 ± 0.02 | 1.37 ± 0.02 | 1.35 ± 0.02 |
| 75 | E-247 | 1147.3 | 3.78 ± 0.14 | 3.77 ± 0.10 | 3.73 ± 0.09 | 3.77 ± 0.10 |
| 76 | E-101 | 1084.7 | 2.40 ± 0.12 | 2.36 ± 0.03 | 2.35 ± 0.08 | 2.41 ± 0.09 |
| 77 | E-271-2 | 1380.0 | 8.85 ± 0.20 | 8.72 ± 0.15 | 8.70 ± 0.13 | 8.60 ± 0.13 |
| 78 | E-249 | 1179.4 | 2.58 ± 0.13 | 2.56 ± 0.10 | 2.53 ± 0.09 | 2.50 ± 0.09 |
| 79 | E-207 | 550.5 | 3.23 ± 0.07 | 3.21 ± 0.05 | 3.17 ± 0.05 | 3.11 ± 0.05 |
| 80 | E-146-2 | 2190.4 | 14.4 ± 0.37 | 14.6 ± 0.29 | 14.7 ± 0.25 | 14.7 ± 0.24 |
| 81 | E-123 | 706.9 | 4.35 ± 0.09 | 4.3 ± 0.07 | 4.32 ± 0.06 | 4.34 ± 0.06 |
| 82 | E-119 | 602.5 | 3.19 ± 0.07 | 3.26 ± 0.06 | 3.26 ± 0.05 | 3.15 ± 0.05 |
| 83 | E-247-3 | 2517.3 | 6.15 ± 0.39 | 6.37 ± 0.29 | 6.27 ± 0.25 | 5.91 ± 0.24 |
| 84 | E-243-1 | 442.7 | 2.42 ± 0.05 | 2.51 ± 0.04 | 2.45 ± 0.04 | 2.50 ± 0.04 |
| 85 | E-100 | 415.1 | 2.61 ± 0.05 | 2.60 ± 0.04 | 2.60 ± 0.04 | 2.60 ± 0.04 |
| 86 | E-285* | 15.5 | 0.079 ± 0.006 | 0.081 ± 0.005 | 0.080 ± 0.004 | 0.083 ± 0.004 |

Table 5 - contd.

| 87 | E-131* | 28.8 | 0.166± 0.010 | 0.167± 0.008 | 0.170± 0.007 | 0.168± 0.006 |
|---|---|---|---|---|---|---|
| 88 | E-275* | 53.6 | 0.086± 0.008 | 0.091± 0.006 | 0.091± 0.006 | 0.093± 0.006 |
| 89 | E-279* | 28.5 | 0.082± 0.007 | 0.141± 0.007 | 0.144± 0.006 | 0.139± 0.006 |
| 90 | E-276* | 35.5 | 0.215± 0.011 | 0.208± 0.009 | 0.208± 0.007 | 0.211± 0.007 |
| 91 | E-278* | 34.1 | 0.172± 0.010 | 0.159± 0.008 | 0.157± 0.006 | 0.157± 0.006 |
| 92 | E-284* | 6.25 | 0.032± 0.004 | 0.030± 0.003 | 0.033± 0.002 | 0.032± 0.002 |
| 93 | E-270* | 5.74 | 0.021± 0.003 | 0.023± 0.002 | 0.027± 0.002 | 0.026± 0.002 |
| 94 | E-200 | 1359.0 | 8.23 ± 0.24 | 8.16 ± 0.18 | 8.37 ± 0.16 | 8.44 ± 0.17 |
| 95 | E-301 | 5360.7 | 44.2 ± 1.6 | 43.7 ± 1.2 | 43.5 ± 0.99 | 43.7 ± 0.88 |
| 96 | E-302 | 159.9 | 0.830± 0.03 | 0.82 ± 0.02 | 0.81 ± 0.02 | 0.80 ± 0.02 |
| 97 | E-303 | 830.1 | 4.66 ± 0.13 | 4.77 ± 0.11 | 4.64 ± 0.09 | 4.55 ± 0.10 |
| 98 | E-304 | 927.7 | 4.52 ± 0.15 | 4.47 ± 0.11 | 4.35 ± 0.10 | 4.57 ± 0.11 |
| 99 | E-305 | 2657.7 | 14.10 ± 0.55 | 14.13 ± 0.42 | 13.9 ± 0.36 | 13.9 ± 0.35 |
| 100 | E-306 | 767.2 | 4.10 ± 0.12 | 4.17 ± 0.10 | 4.19 ± 0.09 | 4.18 ± 0.09 |
| 101 | E-307 | 445.1 | 5.43 ± 0.14 | 5.28 ± 0.11 | 5.24 ± 0.10 | 5.18 ± 0.10 |
| 102 | E-308 | 1875.9 | 10.67 ± 0.35 | 10.79 ± 0.27 | 10.72 ± 0.24 | 10.39 ± 0.24 |
| 103 | E-250 | 22039.1 | 33.67 ± 16.8 | 41.70 ± 10.9 | 45.14 ± 8.1 | 43.82 ± 7.0 |
| 104 | E-208-1 | 1826.8 | 10.16 ± 0.30 | 10.37 ± 0.26 | 10.42 ± 0.23 | 10.76 ± 0.23 |
| 105 | E-271-2 | 1402.2 | 8.69 ± 0.25 | 8.74 ± 0.19 | 9.02 ± 0.17 | 9.31 ± 0.18 |
| 106 | E-255-2 | 2611.6 | 7.59 ± 0.50 | 7.78 ± 0.37 | 7.49 ± 0.82 | 7.35 ± 0.32 |
| 107 | E-269-1 | 4926.2 | 31.39 ± 1.38 | 31.19 ± 1.03 | 31.34 ± 0.86 | 30.52 ± 0.79 |
| 108 | E-281-2 | 8702.0 | 35.62 ± 3.22 | 36.92 ± 2.30 | 36.82 ± 1.82 | 38.17 ± 1.59 |
| 109 | E-137 | 943.4 | 5.34 ± 0.15 | 5.31 ± 0.12 | 5.30 ± 0.11 | 5.33 ± 0.11 |

Table 5 - contd.

| 110 | E-183-2 | 6619.3 | 30.03 ± 2.1 | 31.1 ± 1.5 | 30.7 ± 1.2 | 29.8 ± 1.1 |
|-----|---------|--------|-------------|------------|------------|------------|
| 111 | E-174 | 1439.6 | 6.87 ± 0.24 | 6.81 ± 0.19 | 7.05 ± 0.17 | 6.97 ± 0.17 |
| 112 | E-272-2 | 22346.5 | 49.13 ± 15.8 | 52.7 ± 10.4 | 54.8 ± 7.8 | 48.4 ± 6.9 |
| 113 | E-244-3 | 1948.7 | 9.79 ± 0.36 | 9.70 ± 0.28 | 9.92 ± 0.24 | 10.07 ± 0.24 |
| 114 | E-243-3 | 12947.3 | 17.06 ± 6.4 | 18.8 ± 4.4 | 19.5 ± 3.4 | 20.9 ± 2.9 |
| 115 | E-283-2 | 3176.1 | 7.37 ± 0.65 | 7.24 ± 0.48 | 6.99 ± 0.40 | 6.65 ± 0.39 |
| 116 | E-245 | 4845.5 | 19.1 ± 1.2 | 23.4 ± 0.97 | 18.2 ± 0.78 | 18.8 ± 0.72 |
| 117 | E-250 | 22100.5 | 44.9 ± 15.9 | 48.8 ± 10.5 | 49.0 ± 7.9 | 51.6 ± 6.7 |
| 118 | E-272-2 | 22401.8 | 44.5 ± 16.3 | 50.0 ± 10.7 | 50.3 ± 8.0 | 45.0 ± 7.1 |
| 119 | E-243-3 | 12944.2 | 17.8 ± 6.4 | 20.0 ± 4.4 | 21.3 ± 3.4 | 22.5 ± 2.9 |
| 120 | E-281-2 | 8720.3 | 34.4 ± 3.2 | 34.8 ± 2.3 | 35.3 ± 1.8 | 36.9 ± 1.6 |
| 121 | E-244-3 | 1959.3 | 9.68 ± 0.36 | 9.78 ± 0.28 | 9.80 ± 0.24 | 9.62 ± 0.24 |
| 122 | E-255-2 | 2626.4 | 7.19 ± 0.50 | 7.36 ± 0.37 | 7.41 ± 0.32 | 7.30 ± 0.32 |
| 123 | E-245 | 4877.9 | 17.3 ± 1.3 | 17.8 ± 0.94 | 17.7 ± 0.78 | 17.8 ± 0.72 |
| 124 | E-283-2 | 3143.1 | 7.63 ± 0.64 | 7.20 ± 0.47 | 7.84 ± 0.40 | 7.68 ± 0.39 |
| 125 | E-137 | 948.9 | 5.26 ± 0.15 | 5.29 ± 0.12 | 5.12 ± 0.11 | 5.19 ± 0.11 |
| 126 | E-243-2 | 3425.9 | 6.13 ± 0.71 | 6.64 ± 0.52 | 6.68 ± 0.44 | 5.83 ± 0.43 |
| 127 | E-277-2 | 4374.6 | 9.60 ± 1.05 | 9.87 ± 0.76 | 9.92 ± 0.63 | 9.98 ± 0.60 |
| 128 | E-183-2 | 6670.2 | 29.0 ± 2.1 | 28.8 ± 1.5 | 30.1 ± 1.25 | 28.7 ± 1.1 |
| 129 | E-271-2 | 1399.3 | 8.73 ± 0.25 | 8.76 ± 0.19 | 9.00 ± 0.17 | 9.16 ± 0.17 |
| 130 | E-208-1 | 1819.1 | 10.2 ± 0.33 | 10.4 ± 0.26 | 10.6 ± 0.23 | 10.6 ± 0.23 |
| 131 | E-269-1 | 4923.0 | 29.7 ± 1.37 | 30.7 ± 1.03 | 30.6 ± 0.85 | 30.2 ± 0.77 |
| 132 | E-174 | 1437.3 | 6.76 ± 0.24 | 6.95 ± 0.19 | 6.91 ± 0.17 | 7.16 ± 0.17 |

Table 5 - contd.

| 133 | E-301 | 5442.7 | 46.0 ± 1.62 | 45.0 ± 1.22 | 44.8 ± 1.01 | 44.6 ± 0.90 |
| 134 | E-305 | 2702.4 | 14.6 ± 0.56 | 14.4 ± 0.43 | 14.2 ± 0.37 | 14.1 ± 0.36 |

E 250 gave a count rate of $2.2 \times 10^4$ cps and the ratio of correlated

# 7. FIGURES
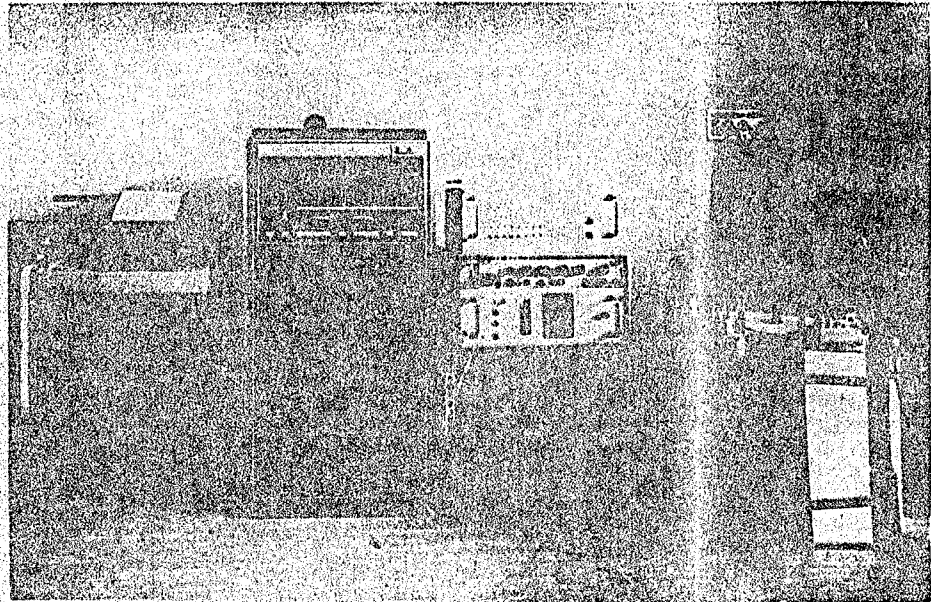


Fig. 1 - GENERAL VIEW OF THE EQUIPMENT
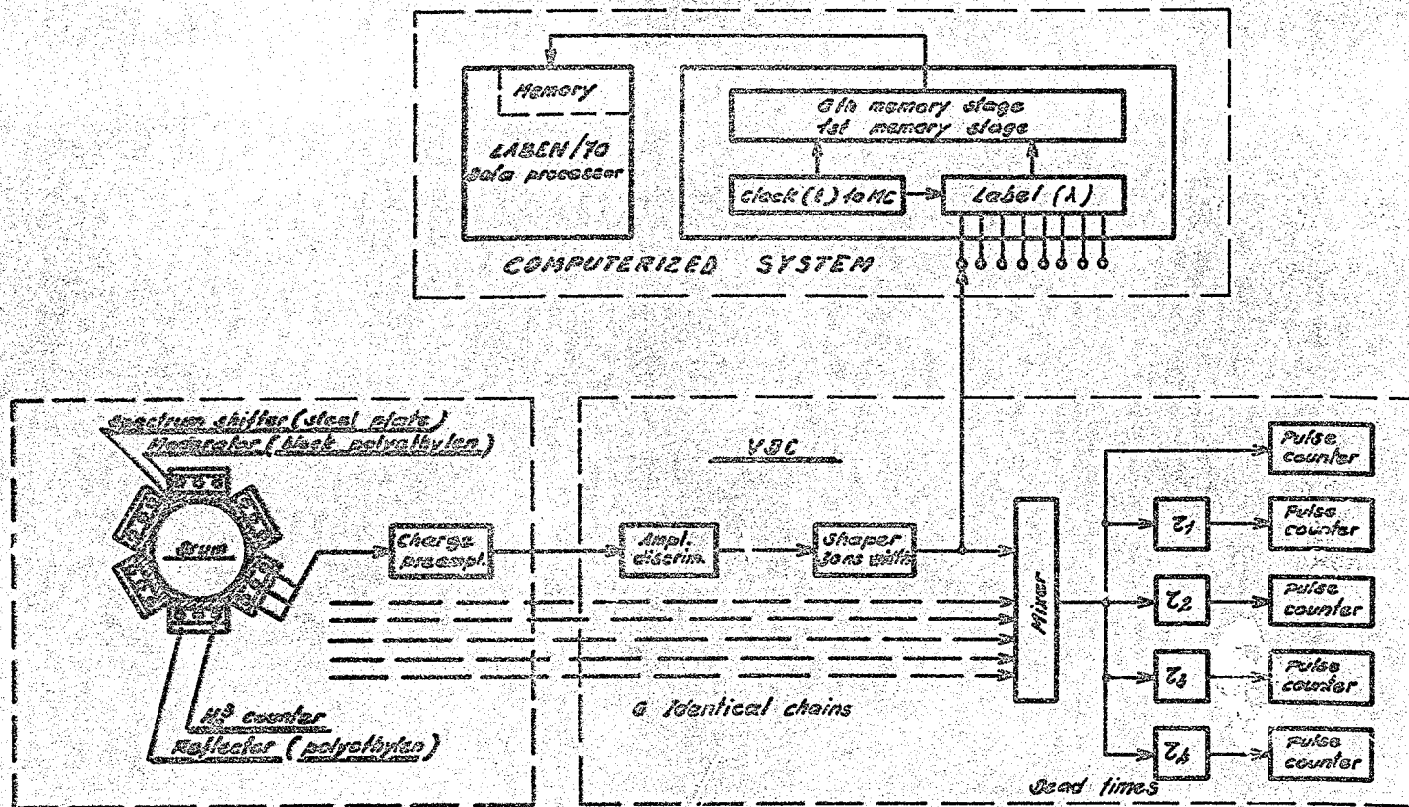
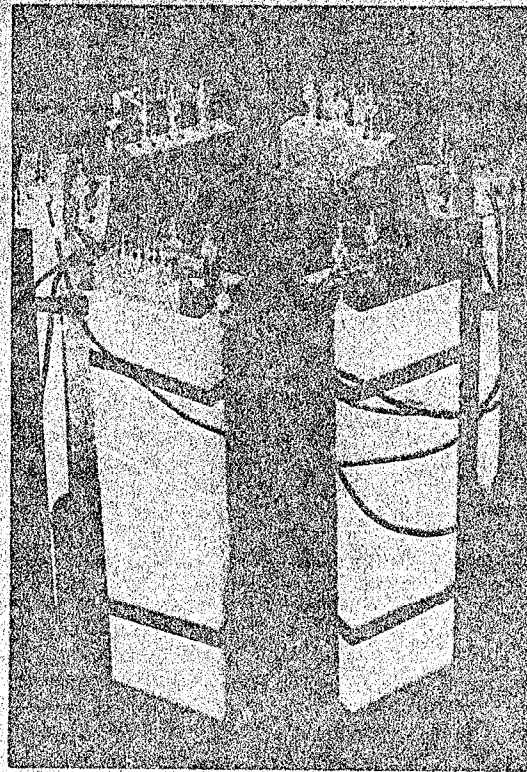Fig. 2 - Block diagram of Euratom passive neutron assay equipment
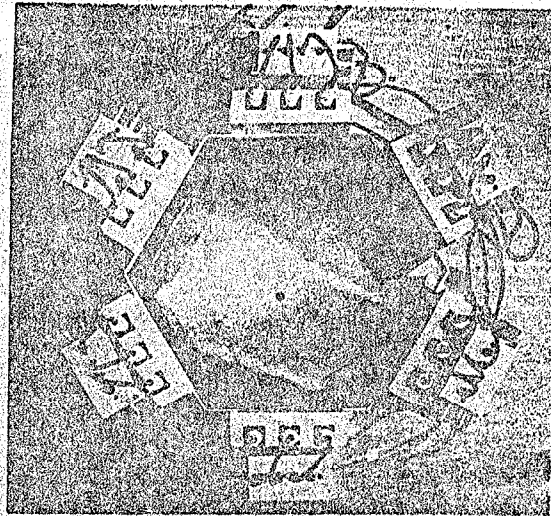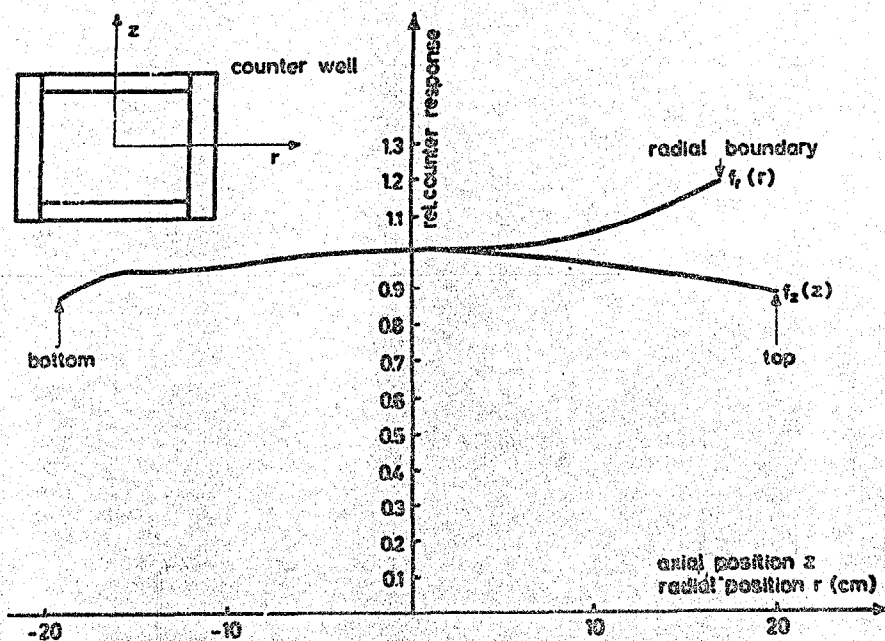
Fig. 3 - TOP AND SIDE VIEWS OF DETECTOR ASSEMBLY

FIG. 4 – AXIAL AND RADIAL COUNTER RESPONSE FUNCTIONS



$$\overline{f_r}(R) = \frac{2}{R^2} \int_0^R f_r(r)\, r\, dr$$

$$\overline{f_z}(H/2) = \frac{1}{H} \int_{-H/2}^{+H/2} f_z(z)\, dz$$

$$\overline{f}(R, H/2) = \overline{f_r}(R)\, \overline{f_z}(H/2)$$
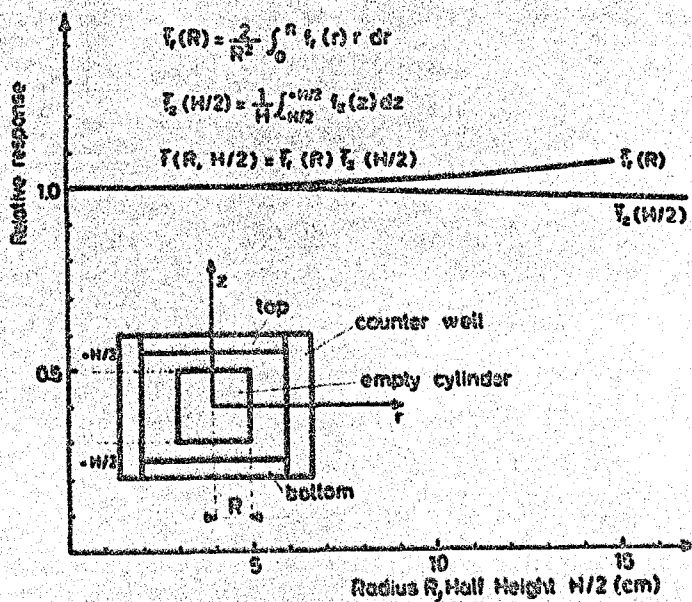
FIG. 5 – COUNTER RESPONSE FUNCTIONS AVERAGED OVER RADIUS R AND HEIGHT H OF EMPTY CILINDRICAL DRUMS

$f(r,z) = f_r(r) \cdot f_a(z); \; f_r(0) = f_a(0) = 1$

$f_r(R) = \frac{2}{R^2} \int_0^R f_r(r) \, r \, dr$

$f_a(R) = \left\{ \frac{1}{H} \int_{-H/2}^{+H/2} f_a(z) \, dz \right\} R$

$c_0(R) = $ counting efficiency in center $(z=0, r=0)$ of drum

counter well

drum of 38cm height filled with polyethylen grains of 0.6 g/cm³ density
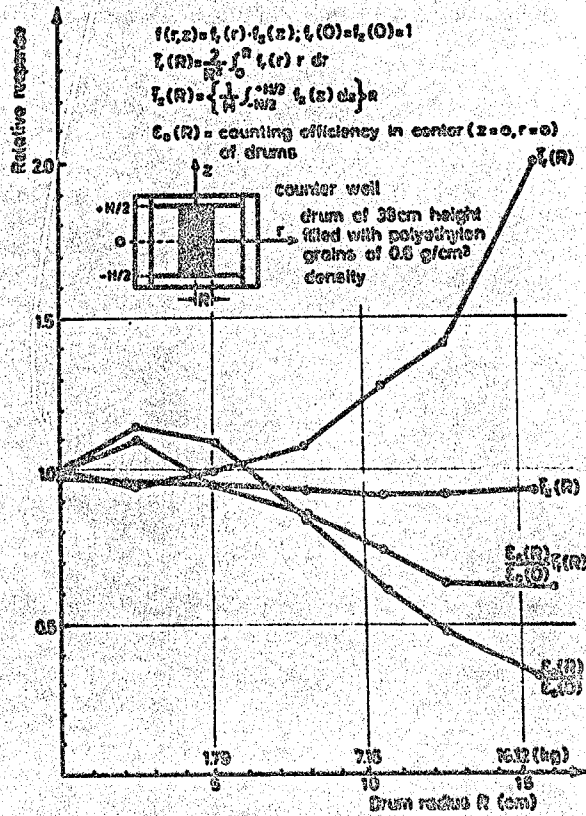
FIG. 6-MODERATING MATRIX EFFECT ON COUNTER RESPONSE FUNCTIONS
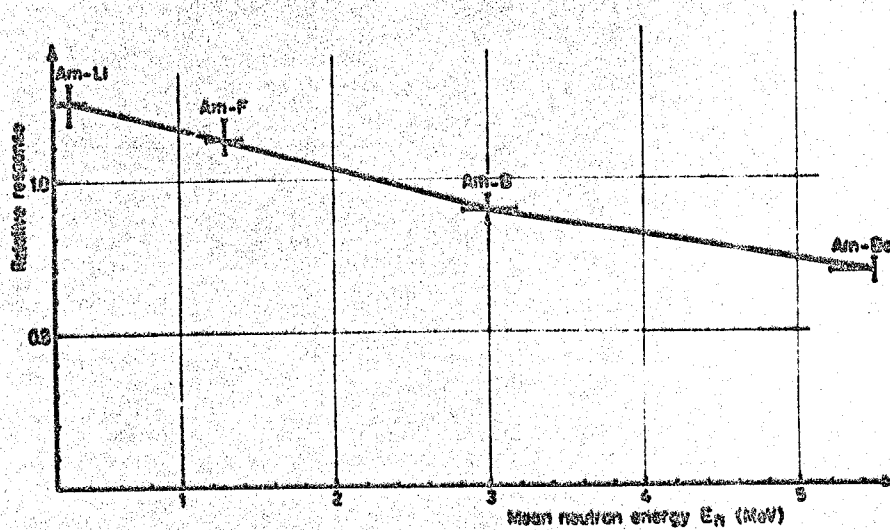
FIG. 7-COUNTER RESPONSE AS A FUNCTION OF NEUTRON ENERGY

VDC - well counter

$$F_i = F_\infty (1 - e^{-T_i/t})$$

FIG. 8 - DETERMINATION OF THE DIEAWAY TIME BY VDC



Detector bias voltage 1030 V
VDC - unit  CC N 2
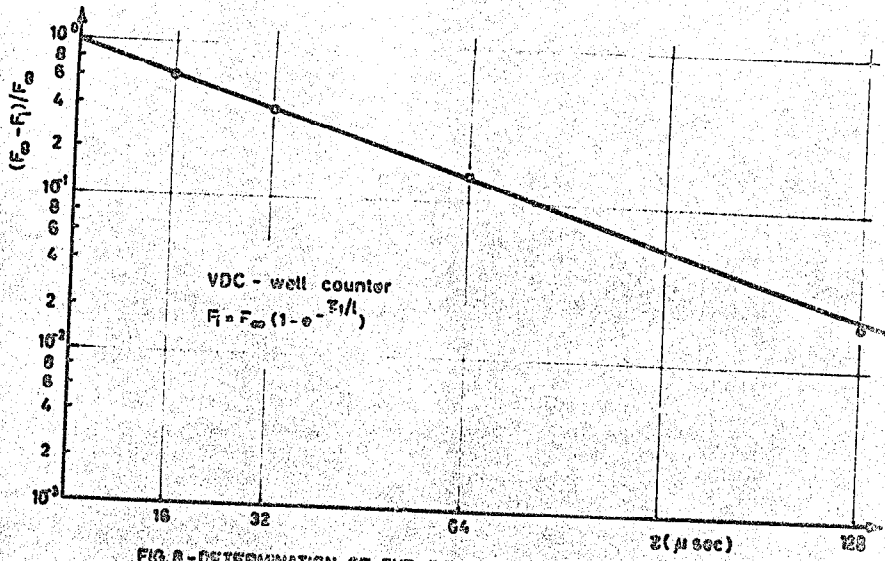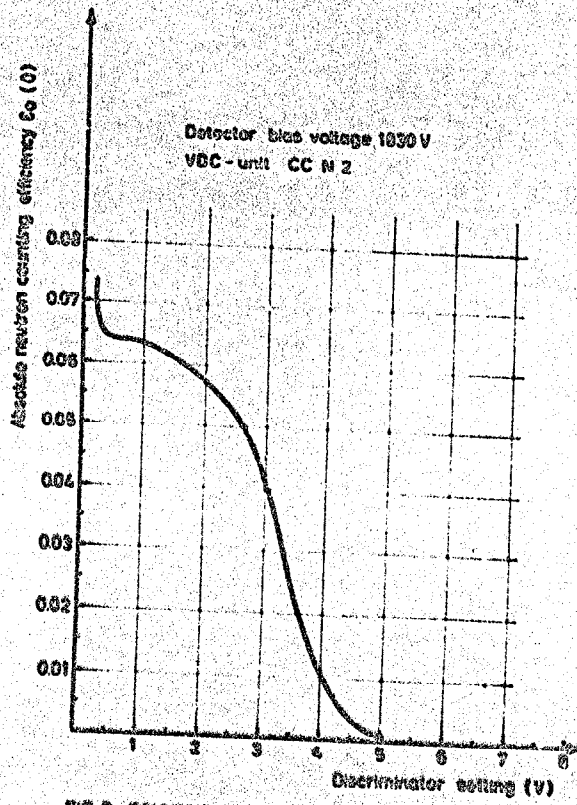
FIG. 9 - COUNTING EFFICIENCY AS A FUNCTION OF
DISCRIMINATOR SETTING

NEUTRON COINCIDENCES COUNTER MOD. CCN 2
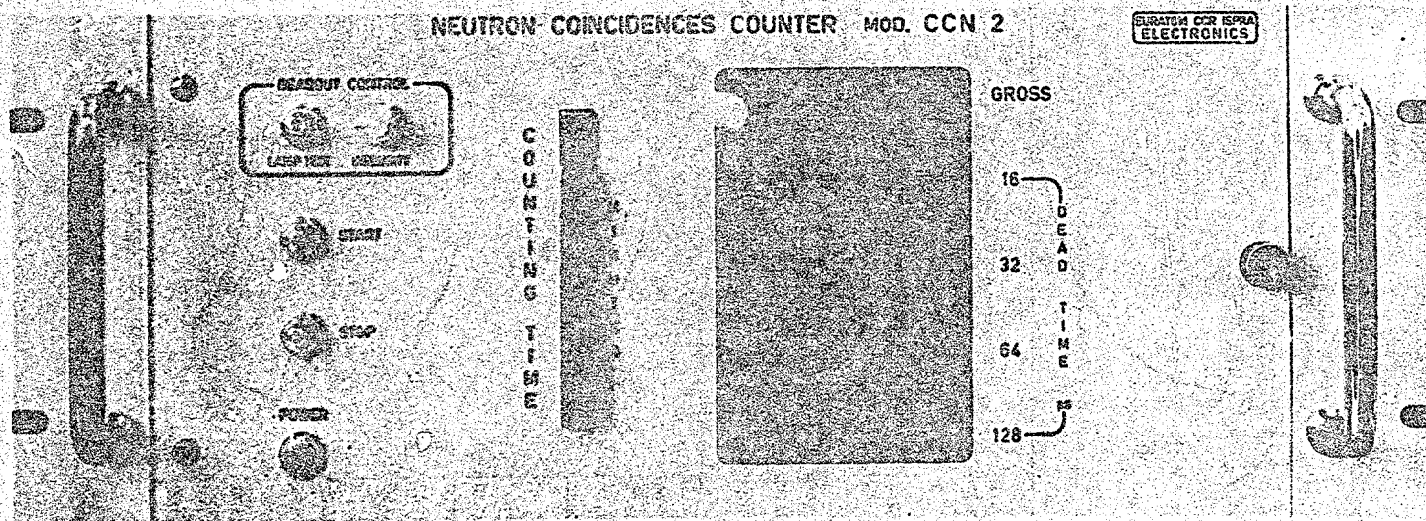
Fig. 10 - THE VARIABLE DEAD TIME (VDC) - COUNTER
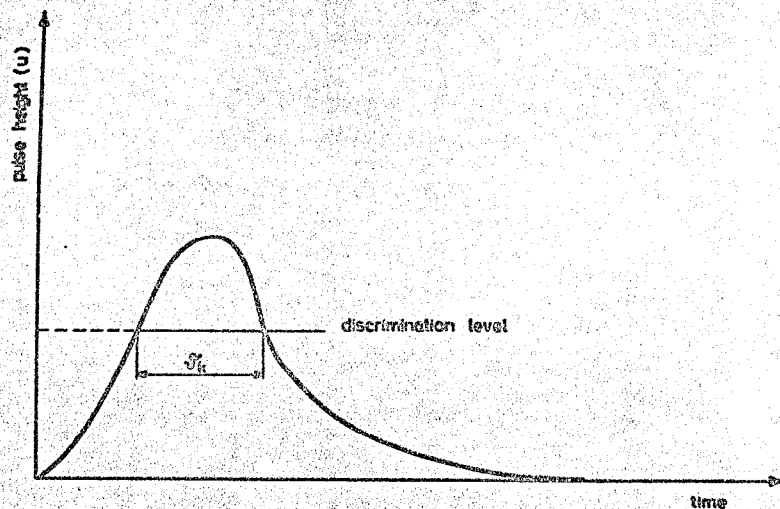
FIG. 11 - EXPLANATION OF THE DISCRIMINATOR PARALYSIS TIME $\vartheta_R$



VDC with 6 pulse forming chains
HV = 1030 V
Cf 252 source in center of the well
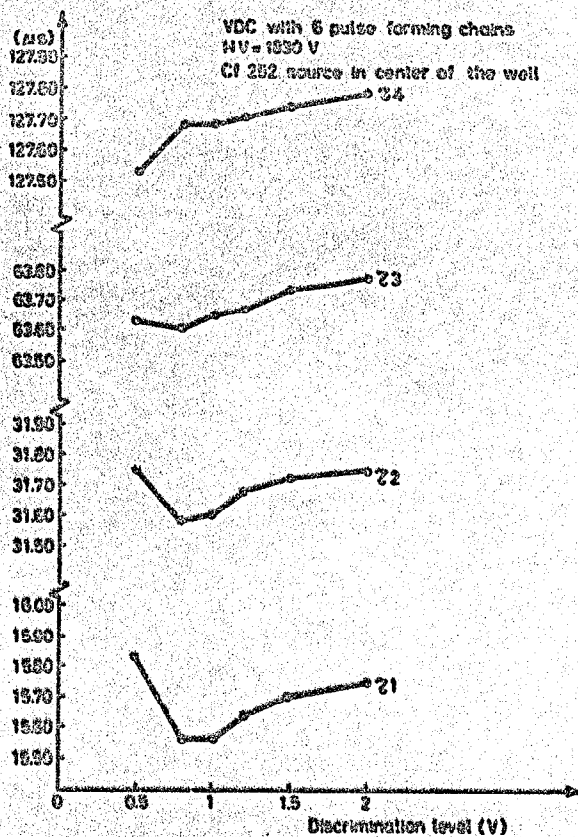
FIG. 12 - COUNTER DEAD TIMES AS FUNCTIONS OF DISCRIMINATION
LEVEL

$$X_{iO} = Xi \ (1 + b_i \cdot C_i)$$

FIG. 13-LINEAR CORRECTION FACTORS (bi) AS FUNCTIONS OF
THE NEUTRON DECAY TIME (t)
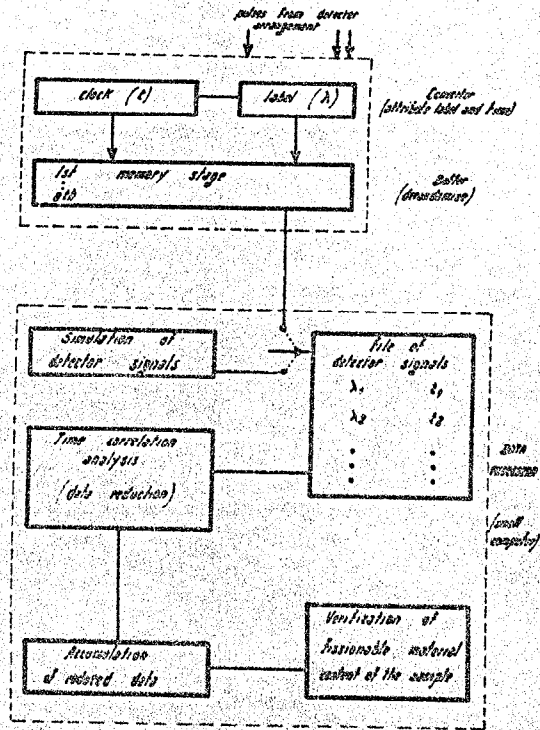
Fig. 19 : Computerized system for time correlation analysis of neutron emission points. (schematic)
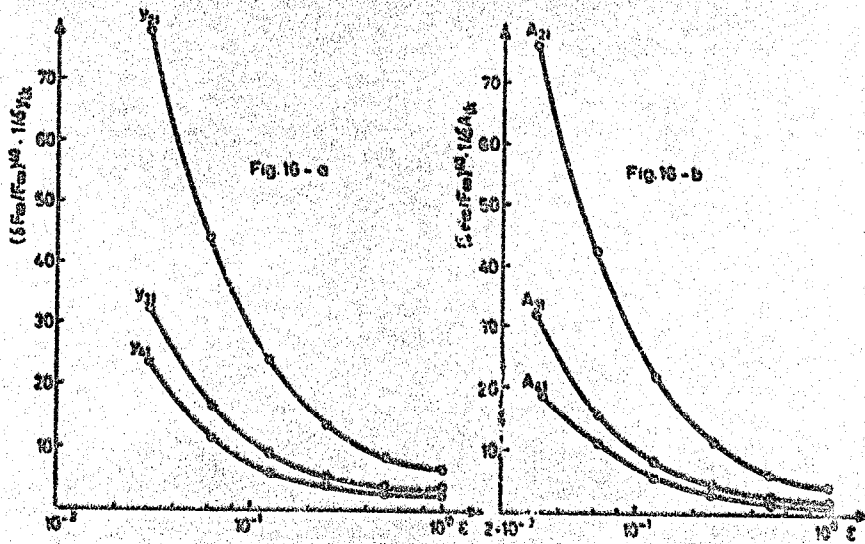


FIG 18-RELATIVE UNCERTAINTY OF $F_{49}$ DETERMINATION AS A FUNCTION OF COUNTING EFFICIENCY $\varepsilon$
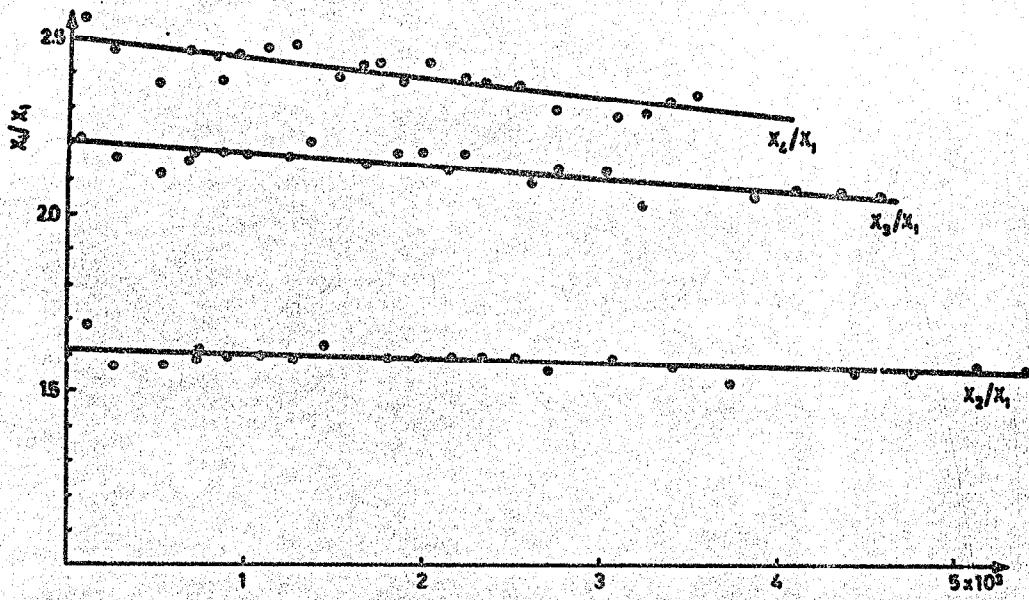
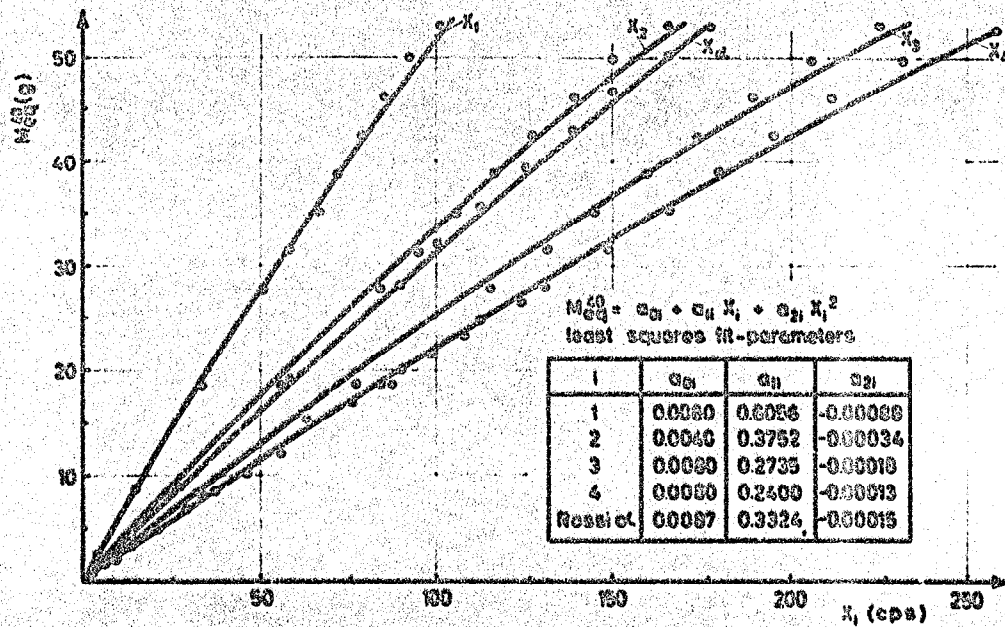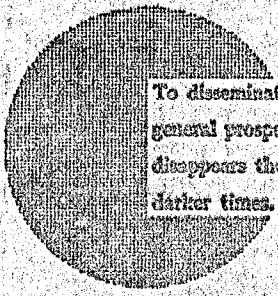FIG. 17—LINEAR VARIATION OF $X_i/X_1$ (i=2,3,4) AS A FUNCTION OF COUNT RATE $C_i$

$C_i$ (cps)



$$M_{eq}^{40} = a_{0i} + a_{1i} X_i + a_{2i} X_i^2$$

least squares fit-parameters

| i | $a_{0i}$ | $a_{1i}$ | $a_{2i}$ |
|---|---|---|---|
| 1 | 0.0080 | 0.6086 | -0.00086 |
| 2 | 0.0040 | 0.3752 | -0.00034 |
| 3 | 0.0080 | 0.2735 | -0.00018 |
| 4 | 0.0080 | 0.2400 | -0.00013 |
| Rossi α | 0.0087 | 0.3324 | -0.00015 |

Fig. 18—CALIBRATION CURVES—VDC ($X_i$)   ROSSI α($X_α$)