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Eine Meßanlage zur Bestimmung von Plutonium in 200 l Abfallfässern

Zusammenfassung

Es wird eine Meßanlage zur Bestimmung von Plutonium in 200 l Abfallfässern der ADB des Kernforschungszentrums Karlsruhe beschrieben. Das Verfahren basiert auf der Zählung von Neutronen. Dazu ist das Faß allseits mit BF_3 - Zählrohren umgeben, die in Polyäthylen eingebettet sind.

Es wurden Messungen an simuliertem Abfall durchgeführt, um den Einfluß von Matrix-Material und einer inhomogenen Plutonium-Verteilung im Faß zu untersuchen. Durch separate Zählratenbestimmung für die äußeren und inneren Zählrohre in einem Moderatorring, der das Faß umgibt, und durch eine damit gewonnene Korrektur konnte die Variation der Nachweisempfindlichkeit von 28% auf 10% reduziert werden. Das Verfahren ist der Messung mit einer Zusatzquelle überlegen. Die Koinzidenzzählrate zeigte eine größere Variation nämlich 18%.

Summary

A neutron well counter is briefly described which will be used for monitoring the plutonium content of 200 l barrels in the waste treatment plant of the Kernforschungszentrum Karlsruhe. Measurements on simulated waste were made to study the influence of matrix material and non-homogeneous plutonium distribution. The variation in detection efficiency could be reduced from 28% to 10% when the signals from inner and outer neutron detectors in the polyethylene annulus are counted separately and a correction is applied, using this information. This method is superior to the source addition technique. Coincidence counting shows a larger variation which could not be reduced to below 18%.

1. Introduction

Plutonium assay by passive neutron counting with a well counter is a well known safeguards technique, both gross neutron counting and coincidence counting being used, (1). Nevertheless each new system and application requires new investigations on the performance characteristics and there is still room for improvements.

In order to validate the observation of German waste storage regulations such a system is now being installed at the KFK radioactive waste treatment plant ADB. Its main objective is the determination of plutonium in waste from the fuel fabrication plant ALKEM, some low level wastes from the reprocessing plant WAK also will be measured. But the majority of the waste from the reprocessing plant is in liquid or homogeneous solid form and is assayed by sampling and chemical analysis. The waste to be measured with the well counter is expected to contain hydrogeneous material in plastic bags loaded into 200 l barrels. But some scrap and non-hydrogeneous material will probably also be present. The throughput of the plant allows 10 minutes counting time per barrel. The detection sensitivity shall be better than 1 g plutonium per barrel. The measurements will be used to categorize the waste drums and to reject those with a too high plutonium content.

For this purpose gross neutron counting will be adequate in most cases: If the counting rate is very low the barrels can pass with the indication that the plutonium content is below a certain threshold. If the counting rate is very high the barrels are rejected and have to be reloaded. For intermediate counting rates coincidence counting will be used to eliminate the (α, n)-contribution.

2. Description of the Facility

The facility essentially consists of a polyethylene annulus of 800 mm inner and 1200 mm outer diameter with a height of 1100 mm. In this annulus two rings of 40 mm diameter, 1000 mm active length BF_3 counters are embedded, each ring consisting of 36 counters. Fig. 1 shows the arrangement of the counters in the annulus. The inner ring has a minimum of 15 mm polyethylene between the counter and the inner surface, for the outer ring this value is 83 mm. In addition top and bottom are loaded with BF_3 counters. The bottom is a polyethlene plate of 160 mm thickness which containes 9 40mm diameter BF_3 counters of 800 mm active length. They are separated by 90 mm from each other and the axes of the counters are 40 mm below the bottom. Holes for insertion of 14 counters are foreseen. Fig. 2 shows the complete facility.

The barrels are loaded into the well from the top and are lowered by means of a hydraulic piston which penetrates the bottom of the well.

The top is covered by a movable polyethylene block rolling on plastic wheels. In this top 9 40 mm diameter 800 mm active length BF_3 counters are embedded. All counters are from the Berthold Comp. and have a filling pressure of 1 Atm. The lower side of the top is covered by a 64 mm lead shield. Then follows a 200 mm polyethylene part in which the counters are embedded, thereafter comes 150 mm solid polyethylene.

Though the facility will primarily be used for low active waste a 70 mm thick lead annulus can be inserted into the well to protect the BF_3-counters from γ-radiation (see Fig. 2). This mode allows operation with activities up to 100 Ci in the barrel.

In order to minimize the background the well counter is located in an underground cavity. It is protected at the top by a 270 mm concrete shield.

In view of the relatively low counting rates of a waste monitor the exit leads of 9 counters are joint together, are fed to one amplifier and are supplied by one high voltage unit. This leads to 8 counter groups from the annulus, one from the bottom part and one from the top part. The 8 groups in the annulus correspond to 4 90^{o} sectors and in each sector the inner and the outer counters are connected separately. In addition the total counting rate of all detectors is measured.

Plateau curves were measured for each counter and those with similar characteristics were grouped together in order to maintain a certain plateau even when 9 counters are connected in parallel.

For coincidence counting the system described in (1) is used. All counting rate data are printed on paper tape for manual processing. A computerized data aquisition system is foreseen for routine operation in the future.

An automatic barrel transport system and tagging of the barrels is also planned. The time of implementation is coupled with construction work at the ADB site as well as development and delivery times of the components.

3. Investigations on Matrix and Plutonium Distribution Effects

Before the facility was brought to the KFK waste treatment plant, proper functioning was checked and measurements were made in the laboratory on simulated waste drums to study the influence of matrix material and fuel distribution in the drum.

The simulated waste consisted of polyethylene tube and rod pieces the dimensions of which were selected to give average densities of $\rho = 0.2\ g/cm^3$ and $\rho = 0.3\ g/cm^3$ when filled into the barrel. In addition measurements were made on an empty barrel. This covers the expected range of hydrogen densities in the real waste barrels, $\rho = 0.3\ g/cm^3$ being already some what high.

The barrel had - like the German standard - an inner diameter of 560 mm and the filling height was about 800 mm. To simulate the density of $\rho = 0.2\ g/cm^3$ polyethylene tubes of 32 mm outer diameter and 3 mm wall thickness were used. The length of the tubes was 100 mm. The density of $0.3\ g/cm^3$ was obtained by mixing 32 kg of polyethylene rods of 20 mm diameter with 28 kg of tubes of 25 mm outer diameter and 2 mm wall thickness, the length being always 100 mm.

No plutonium probes of adequate neutron emission rates for parametric studies were available. Therefore a ^{252}Cf source was used to simulate plutonium. For gross neutron counting only the difference in the fission neutron spectra is relevant, with the mean neutron energy being 13 % higher for ^{252}Cf than for ^{240}Pu(1). This leads to a 4 % larger slowing down distance in water or polyethylene. In the barrel and well the neutrons travel roughly one slowing down length, thus the results of these measurements may be some per cent different from what would be obtained with ^{240}Pu. The conclusions probably remain unaffected. The difference is much more important for coincidence counting as will be explained later.

For positioning of the source in the barrel a 10 mm diameter tube was inserted vertically at the following 4 radial distances from the barrel axis: 0.0, 8.75 cm, 17.4 cm, and 26.0 cm. At each radial distance measurements were made at 8 vertical source positions, the distance between positions being 10 cm. The measurements were repeated for different angles between source and barrel covering 180° in 15° steps. In view of the 4 sector symmetry of the system only a range of 45° was measured. For the empty barrel 90° were covered to check symmetry and to gain redundancy.

Fig. 3 shows for illustration the variation of the counting rate in one detector group with the angle ϕ between source and detector bank center, ϕ is measured from the barrel axis. The strong angular dependence clearly indicates that each detector group essentially counts the sources in front of it, what allows to detect inhomogeneities in the plutonium or matrix material distribution.

Tables 1 to 3 show for the 3 densities the total counting rate of all detectors including top and bottom as a function of radial and axial source position. The figures given are ε = counts per source neutron. The source height position h is given in cm above bottom of the barrel. In addition the ratio of inner ring counting rate to outer ring counting rate is given.

4. Evaluation of the Measurements

4.1. Inner - to - Outer Counting Rate Correction

Tables 1, 2 and 3 reveal that there is a large variation in detection sensitivity depending both, on source location and matrix density. The sensitivity increases with the distance of the source from the barrel axis and decreases with increasing matrix density. The average sensitivity is

$\bar{\varepsilon} = 0.11 \pm 28$ % (standard deviation) counts per source neutron.

The ratio of counting rates of the inner ring to the outer ring of counters in the annulus $\frac{I}{O}$ shows a trend in the opposite direction: It increases with the matrix density and decreases with the distance of the source from the barrel axis. Unfortunately the latter effect does not hold for the highest density. Moreover the ratio $\frac{I}{O}$ decreases rapidly for source positions close to the top. Therefore the ratio of counting rates of the detectors in the top cover "T" to the detectors in the bottom "B" also was used for correction. One gets a relatively flat behaviour for $\varepsilon(\frac{I}{O} + 0.05\frac{T}{B})$. Therefore it is recommended to determine the source intensity s from the total counting rate c_{tot} by

$$s = \frac{c_{tot}}{\varepsilon} = \frac{c_{tot}}{\varepsilon(\frac{I}{O} + 0.05\frac{T}{B})} \quad (\frac{I}{O} + 0.05\frac{T}{B})$$

$$s = 3.25\ c_{tot}(\frac{I}{O} + 0.05\frac{T}{B}) \pm 10\ \%$$

(1) s = source intensity (n/sec)

c_{tot} = total counting rate

I = counting rate of inner ring

O = counting rate of outer ring

T = counting rate of top cover counters

B = counting rate of bottom counters

The correction reduces the standard deviation from 28 % to 10 % without the requirement of an additional measurement.

This variation is pessimistic insofar as it assumes that the neutron sources in the barrel are concentrated in one point, whereas in reality they will be more distributed. For the other extrem of homogeneously distributed sources the sensitivity ε increases when going from $\rho = 0.3\ g/cm^3$ to $\rho = 0.0\ g/cm^3$ by a factor of 1.58. For $\varepsilon(\frac{I}{O} + 0.05\frac{T}{B})$ the factor is only 1.10.

4.2. Source Addition Technique

To reduce the effect of matrix material on detection sensitivity a so called "source addition technique" was proposed by H.O. Menlove and R.B. Walton (2). It works as follows: A small spontaneous fission source is positioned at the surface of the barrel and the counting rate of the well counter resulting from this source is determined. Then the counting rate of the barrel without source is normalized to that from the source. In this way the matrix effect is eliminated or at least significantly reduced since both counting rates respond in a similar way on the matrix material.

This method also has been used here by positioning the ^{252}Cf-source at the barrel surface in the barrel midplane symmetric to one group of counters. The response of the following detector groupings was determined:

1. all counters including top and bottom
2. the detector groups in the annulus (inner + outer) at 90^o to the source
3. the detector groups in the annulus (inner + outer) opposite to the source.

For a homogeneous distribution of fissile material in the barrel - which is obtained by integration over the point source measurements - the best results were obtained for the 90^o detectors. They are summarized in Table 4.

The variation in counting efficiency $(\frac{Max}{Min})$ is reduced from a factor of 1.58 to 1.06 by the source addition technique. However, by virtue of its nature, this method can only correct for the matrix density but not for space dependence effects of the source distribution in the barrel. Thus, for the case that all fissile material in the barrel is concentrated at one point

Table 4 The Effect of Source Addition Normalization on Homogeneously Filled Barrels, Detectors in 90° Angle to the Source

ρ (g/cm^3)	ε	ε_s	$\frac{\varepsilon}{\varepsilon_s}$
0.0	0.1384	$2.77 \cdot 10^{-2}$	5.00
0.2	0.1075	$2.06 \cdot 10^{-2}$	5.22
0.3	0.0875	$1.78 \cdot 10^{-2}$	4.92

ε = well counter efficiency to sources in the barrel (counts per source neutron)

ε_s = 90° detector efficiency to external source (counts per source neutron) the correction is less efficient. It reduces the standard deviation of ε from $\pm$ 28 % to $\pm$ 15 % whereas the method described above gives a reduction to $\pm$ 10 %. In order to get more experience both methods will be used and compared on actual waste drums. The source addition technique has of course the disadvantage of requiring an additional measurement.

5. Coincidence Counting

Neutrons are in waste barrels not only emitted by spontaneous fission processes but also result from (α, n)-reactions on oxygen or flourine with α's from plutonium or the α-decay of other actinides. The main α-emitter is often ^{238}Pu, the isotopic content of which is generally not well known, so that these neutrons can not be used for a plutonium assay. They can be eliminated by an electronic logic which makes use of the fact that fission neutrons are emitted in pairs thus causing detector signals which are correlated in time whereas the (α, n)-neutrons are uncorrelated. For details the reader is referred to (1).

The counting rate of the correlated signals is approximately given by:

(2) $$C_c(\Delta t) = \varepsilon^2 \frac{\overline{\nu(\nu-1)}}{2} (1-e^{-\alpha\Delta t}) \cdot sf$$

where:

$\frac{1}{\alpha}$ = neutron life time in the point model approximation (exponential decay as a response to a neutron injection).

Δt = coincidence interval (= gate width)

$\frac{\overline{\nu(\nu-1)}}{2}$ = average number of neutron pairs emitted per fission = 5.7 for ^{252}Cf
= 1.87 for ^{240}Pu

sf = fission rate

$C_c(\Delta t)$ being proportional to ε^2 the variation with source position and matrix density is about twice as large as for the gross counting rate. In addition α varies with the matrix density: since neutron lifetime is increasing with increasing hydrogen density $(1-e^{-\alpha\Delta t})$ is decreasing. Thus, it has the same tendency as has ε.

In Table 5 the correlated signal as calculated from Equ.(2) is compared with the measured values. In the calculation ε-values as determined from the gross counting rate are used. For α a constant value of $\frac{1}{\alpha} = 80$ µsec was taken, which has been measured at the empty barrel. The dependence on matrix density and radial source position, axially averaged, are given. The deviations between measurement and calculation may partly result from neglecting the dependence of α with hydrogen density but partly they may result from the point model approximation of Equ.(2).

The variation with source position and matrix density of the correlated signals was found to be $\pm$ 62 % (standard deviation). This is highly unsatisfactory. In case that the spatial distribution of fission and (α, n) neutron sources is the same a correction using the gross neutron counting rate can be applied. This is the case when PuO_2 is the only neutron emitter in the barrel, for instance.

Some corrections were tried and the best result was obtained for $C_c \cdot (\frac{I}{O})^3$ where the standard deviation is $\pm$ 18 %. For $C_c(\frac{I}{O} + 0.05\,\frac{T}{B})^3$ a similar variation was found. The notations are explained in paragraph 4.1.

The source addition technique was also applied for the correlated signals. But in contrast to gross neutron counting correlated signals were not available for detector groups but only for all detectors together.

Two evaluations were made: In one case the correlated signal was normalized to the square of the gross counting rate of the detector groups perpendicular to the external source. In the other case the correlated signal from the external source was used for normalization. In the first case a standard deviation of $\pm$ 33 % in the second case of $\pm$ 35 % was found. The remaining large variations result primarily from space dependence effects at the highest matrix density which are notreduced by the source addition technique.

The situation looks entirely different for homogeneous source distributions, which can be obtained by integration over the results from measurements with different source positions. Then the variation of the correlated signal is 1.14 $(\frac{\text{Maximum}}{\text{Minimum}})$ when normalized to the square of the gross counting rate of the 90° angle detectors and 1.57 when normalized to the correlated signal from the external source. The superiority of the 90° position is evident.

Table 5 Comparison of Measured and Calculated Correlated Signals

	R(cm):	0.0	8.75	17.4	26.0
ρ = 0.0 g/cm^3	ε	0.1335	0.1360	0.1379	0.1414
	C_c calc.	$2.64\cdot10^3$	$2.73\cdot10^3$	$2.81\cdot10^3$	$3.29\cdot10^3$
	C_c exp.	$2.6\cdot10^3$	$2.6\cdot10^3$	$2.65\cdot10^3$	$2.65\cdot10^3$
	calc./exp.	1.02	1.05	1.06	1.24
ρ = 0.2 g/cm^3	ε	0.0937	0.0978	0.1058	0.1181
	C_c calc.	$1.29\cdot10^3$	$1.42\cdot10^3$	$1.66\cdot10^3$	$2.06\cdot10^3$
	C_c exp.	$1.0\cdot10^3$	$1.1\cdot10^3$	$1.3\cdot10^3$	$1.57\cdot10^3$
	calc./exp.	1.29	1.29	1.28	1.31
ρ = 0.3 g/cm^3	ε	0.0657	0.0716	0.0838	0.1060
	C_c calc.	$0.64\cdot10^3$	$0.76\cdot10^3$	$1.04\cdot10^3$	$1.66\cdot10^3$
	C_c exp.	$0.5\cdot10^3$	$0.6\cdot10^3$	$0.8\cdot10^3$	$1.45\cdot10^3$
	calc./exp.	1.28	1.27	1.30	1.15

sf = $4.71\cdot10^4$ fissions/sec

C_c = counts per second

Δt = 100 μsec

All results were obtainen with a ^{252}Cf source. But ^{252}Cf emits more neutrons per fission than ^{240}Pu and has a 3 times higher coincidence counting rate for the same fission rate. Thus, verification of the results with ^{240}Pu is important.

6. Summary and Conclusions

Measurements were made to investigate the dependence of the counting rate of a well counter on fissile material distribution and matrix material in the barrel. Polyethylene tubes were used to simulate hydrogeneous waste and the density was varied between 0 and 0.3 g/cm^3. A ^{252}Cf source was used to simulate plutonium and the source position was varied over the whole barrel volume. In order to reduce the counting rate variation the counting rates of an inner and an outer ring of detectors in the polyethylene annulus of the well counter were measured separately and a correction was applied on the basis of this information. Alternatively the "source addition technique" was used to reduce the variation with hydrogen density (2). Both, gross neutron counting and coincidence counting were analyzed. The coincidence measurements were made with the system developed and described by Böhnel (1).

The results are summarized on Table 6. The measurements with single point sources contain data from 32 space points times 3 densities so that the variation of the counting rate could be well analyzed and the standard deviation σ is given in the Table. The data for a homogeneous source distribution are obtained by integration of the space point measurements. Then only 3 values, one for each density, are gained and the standard deviation is not very meaningful any more. Therfore here the ratio of maximum to minimum value is given.

One can draw the following conclusions from the measurements:

1. The variation of the correlated signal is about twice as large as that of the gross counting rate. This is due to the fact that the correlated signal is proportional to the square of the detection efficiency. In addition there enters the variation of neutron life time with barrel fillings.

2. The correction on the basis of inner ring to outer ring counting rate reduces the variation by about a factor of 3.

3. This correction is superior to the source addition technique when the fissile material in the barrel is very inhomogeneously distributed. For

Table 6 Summary of Results from Simulated Waste Measurements Variation of Detector Response with Matrix Density and Fission Source Position

	gross neutron counting		correlated signal	
	point sources 1 standard deviation	homog. sources $\frac{Max}{Min}$	point sources 1 standard deviation	homog. sources $\frac{Max}{Min}$
uncorrected	$\pm$ 28 %	1.58	$\pm$ 62 %	2.79
$(\frac{I}{0})$ correct.	$\pm$ 10 %	1.10	$\pm$ 18 %	1.23
source addition correction	$\pm$ 15 %	1.06	$\pm$ 33 % ($\pm$ 35 %)	1.14 (1.57)

( ) = normalized to correlated signal from an external source homogenous distribution the effect of the corrections are similar, the source addition being slightly better.

4. A good result for the source addition technique was only observed when a detector group with 90^{o} angle to the external source was used.

5. For an accurate determination of the plutonium content in a barrel the correlated signal has to be used in order to suppress the (α, n)-contribution. But just in this case the variation is high, even after corrections. Thus, satisfactory results with about 5 % standard deviation only will be obtained when the fissile material is fairly homogeneously distributed and the variation in hydrogen density is limited.

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Los Alamos Scientific Laboratory, Nuclear Safeguards Research and Development
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Table 1

Dependence of detection sensitivity ε (counts per source neutron) and inner ring to outer ring counting rate ratio $\frac{I}{0}$ on source location in the barrel. Polyethylene density in the barrel ρ = 0.0 g/cm^3.

ε

h \ R	0.0	8.75	17.4	26.0
90	0.110			
80	0.119	0.123	0.123	0.128
70	0.129	0.132	0.135	0.138
60	0.135	0.139	0.140	0.144
50	0.139	0.140	0.142	0.147
40	0.141	0.143	0.144	0.148
30	0.141	0.142	0.143	0.147
20	0.139	0.140	0.142	0.143
10	0.125	0.129	0.134	0.136
	.1335	.1360	.1379	.1414

$\frac{I}{0}$

h \ R	0.0	8.75	17.4	26.0
90	2.17	2.25	2.30	2.35
80	2.09	2.20	2.27	2.32
70	2.11	2.18	2.24	2.31
60	2.09	2.18	2.22	2.31
50	2.02	2.04	2.11	2.26
40	2.11	2.18	2.24	2.32
30	2.24	2.27	2.31	2.38
20	2.20	2.26	2.33	2.44
10	2.30	2.35	2.39	2.45

bottom

R = distance from the barrel axis in cm

h = distance from the bottom of the barrel in cm

Table 2

Dependence of detection sensitivity ε (counts per source neutron) and inner ring to outer ring counting rate ratio $\frac{I}{0}$ on source location in the barrel. Polyethylene density in the barrel ρ = 0.2 g/cm^3.

h \ R	ε 0.0	ε 8.75	ε 17.4	ε 26.0	I/0 0.0	I/0 8.75	I/0 17.4	I/0 26.0
80	0.0907	.0959	.1011	.1136	2.59	2.50	2.49	2.42
70	0.0935	.0984	.1059	.1214	3.07	3.10	2.89	2.52
60	0.0969	.1013	.1112	.1242	3.22	3.28	2.92	2.62
50	0.0977	.1015	.1118	.1252	3.28	3.34	2.99	2.49
40	0.0962	.1013	.1100	.1217	3.32	3.32	3.04	2.59
30	0.0936	.0976	.1067	.1192	3.35	3.36	3.02	2.49
20	0.0913	.0944	.1024	.1123	3.32	3.35	3.03	2.56
10	0.0894	.0918	.0976	.1069	3.26	3.25	3.05	2.58

R = distance from the barrel axis in cm

h = distance from the bottom of the barrel in cm

Table 3

Dependence of detection sensitivity ε (counts per source neutron) and inner ring to outer ring counting rate ratio $\frac{I}{O}$ on source location in the barrel. Polyethylene density in the barrel $\rho = 0.3$ g/cm^3

h \ R	ε 0.0	ε 8.75	ε 17.4	ε 26.0	I/O 0.0	I/O 8.75	I/O 17.4	I/O 26.0
90	.0676	.0732	.0823	.0949	2.64	2.16	2.18	2.33
80	.0610	.0661	.0783	.1029	3.62	2.51	2.59	2.51
70	.0586	.0641	.0775	.1106	3.99	3.35	3.32	2.71
60	.0637	.0718	.0800	.1133	3.91	3.68	3.56	2.69
50	.0686	.0764	.0869	.1155	3.84	3.58	3.42	2.69
40	.0699	.0761	.0920	.1132	3.77	3.57	3.46	2.65
30	.0680	.0735	.0896	.1088	3.75	3.60	3.41	2.64
20	.0624	.0685	.0842	.1007	3.95	3.62	3.21	2.69
10	.0719	.0746	.0835	.0941	3.67	3.45	3.31	2.82

R = distance from the barrel axis in cm

h = distance from the bottom of the barrel in cm

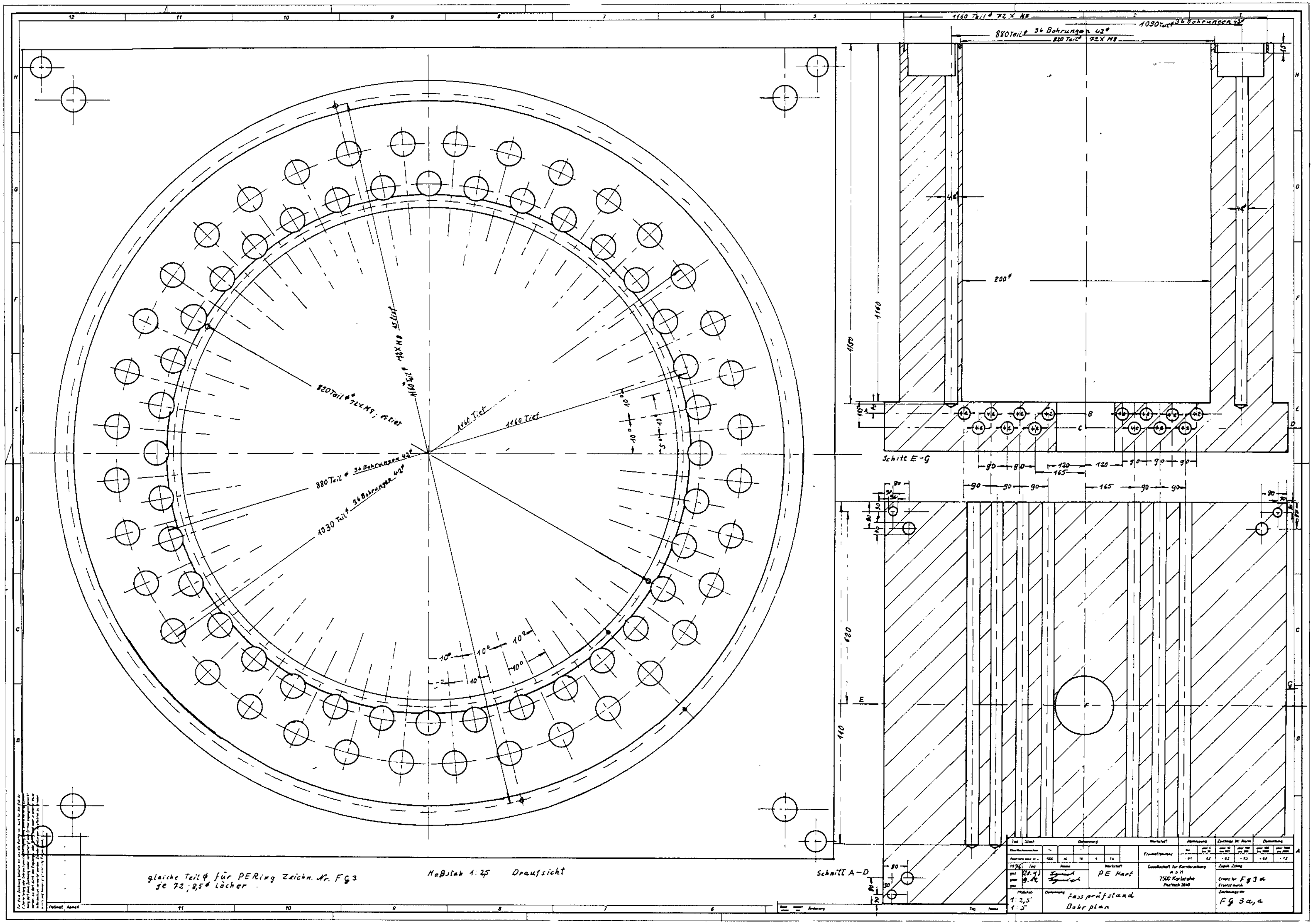

Fig. 1 Arrangement of Counters surrounding the Waste Barrel

Legend to Fig. 2

0 Waste barrel
1 polyethylene well
1a inner ring of counters
1b outer ring of counters
1c holes for bottom counters
2 lead insert
3 base plate, adjustable
4 sliding top cover
4a holes for counters
5 hydraulic piston drive
6 hydraulic cover drive
7 rails for cover
8 rolls for barrel transport
9, 10 movable concrete cover
11 cavity

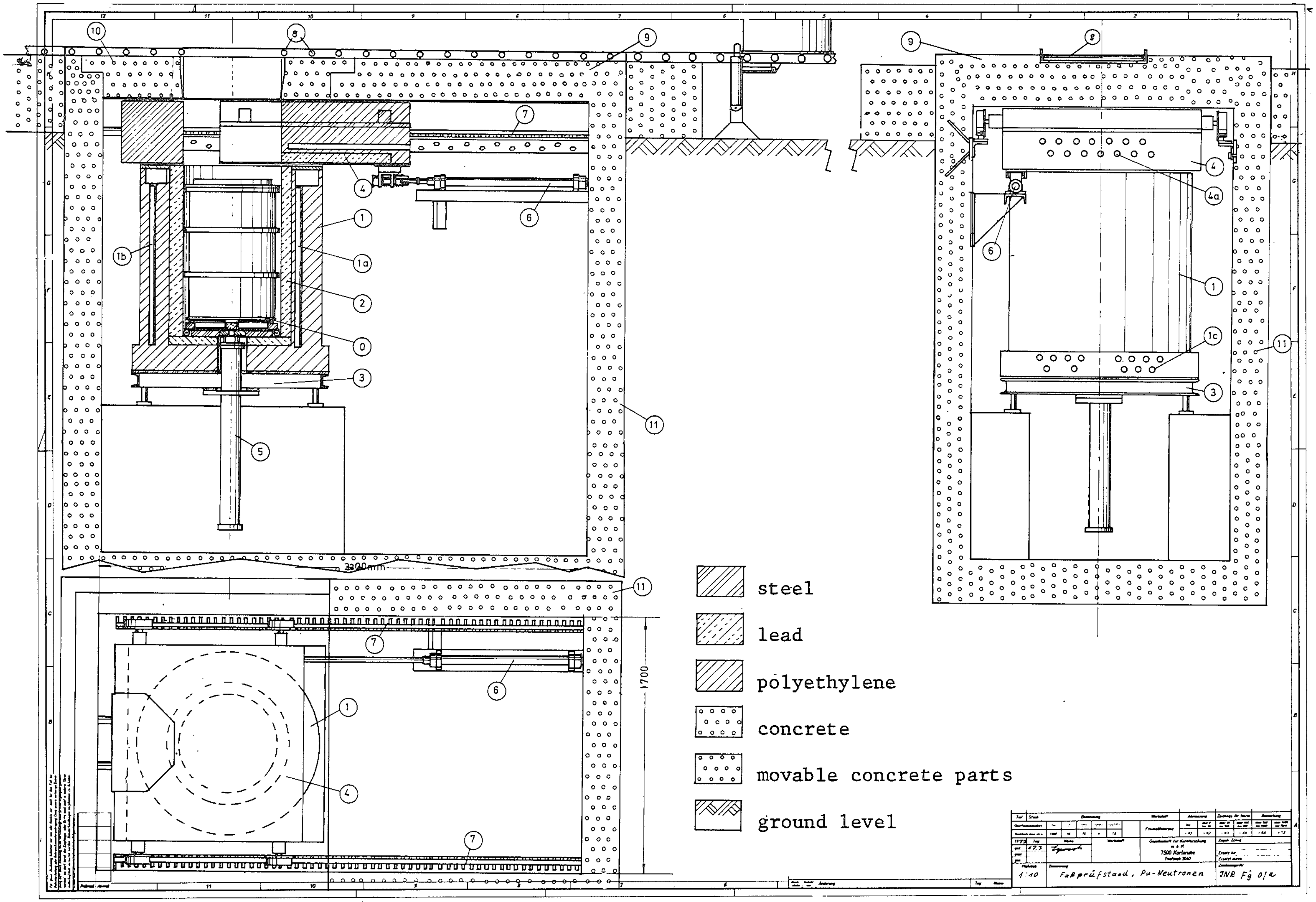

Fig. 2   Neutron Well Counter
For Explanation of Figures see Legend

Fig. 3 Angular Dependence of Counting Rate
Radial Source Distance R as Parameter