

**KERNFORSCHUNGSZENTRUM  
KARLSRUHE**

Februar 1969

KFK 901

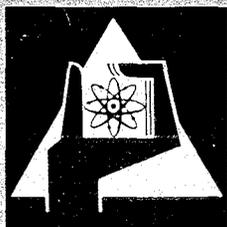
Institut für Angewandte Reaktorphysik

Safeguard Exercise at the Fabrication Plant Alkem

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with the assistance of

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## Introduction

The main objective of a modern safeguards system established in [1] is to warrant a high degree of security against the misuse of fissionable material in the domain of peaceful use of nuclear energy.

In the framework of the "Spaltstoffflußkontrolle (SPFK)" project performed at the Karlsruhe Nuclear Research Center several activities are scheduled in the field of systems analysis and development of instruments to establish such a modern safeguards system.

One of the objectives of this project is the practical application of the theoretical results in the field systems analysis and the demonstration of the instruments developed. For that purpose several safeguards exercises are scheduled in existing plants at different stages of the project.

The first exercise of this type was performed between May and September 1968 at the ALKEM fabrication plant (Alpha-Chemie and Metallurgie) located on the premises of the Karlsruhe Nuclear Research Center. The planning and the execution of this first exercise are described in this paper.

### 1. Planning the Exercise

#### 1.1. Objectives

For application of the principles of the safeguards system described in [1], the first exercise has been carried out at the ALKEM company, where some 200 kg of Pu are processed annually under commercial conditions. The exercise has been carried out for a typical fabrication campaign. A number of problems not considered before were caused by the necessity to perform the exercise in an existing plant. The first problem occurred as a result of the impossibility to identify the strategic points on the basis of theoretical considerations only but instead, they had to be identified according to the possibilities of the plant layout. Secondly, it was necessary to establish a certain set of so-called special measures in addition to the measures considered, such as throughput measurement, inventory taking and containment measures. It became evident that these special measures would not be required or would be redundant in future plants with a layout better suited to the

concept of strategic points. On the other hand, it was possible to demonstrate that the principles of safeguarding the flow of fissionable material at a limited number of strategic points can be applied to existing plants with some minor important additional, special measures being added and without the use of highly automated instruments and techniques.

For the exercise the following objectives were established:

1. For the existing fabrication plant ALKEM, the safeguards exercise was to be carried out for a typical fabrication campaign. Safeguards was defined in this connection to be the establishment of the balance of plutonium with the use of only a small number of strategic points. If possible, only such safeguards measures were to be applied, which could be applied also to future plants, constructed more suitable to the concept of strategic points established in [1], [2], [3]. If, however, special measures should be necessary, it was to be tested, whether these measures were redundant in future plants as a result of the provision of tamperproof instruments and tight containments.
2. With the results of the measurements at the different strategic points and with the results of the calculated material balance the statements developed in [1] from a more theoretical point of view were to be corroborated in the exercise. These statements to be established from the results of the exercise were the probability of diversion and the risk of detection. To demonstrate firstly the validity of these statements, the management of ALKEM was to try to withdraw a certain amount of material from the stream of plutonium during the campaign that was safeguarded. To make secondly the exercise more realistic, the fact of this withdrawal and the amount withdrawn, unknown to the safeguard people, was to be detected only by the methods of the material balance.
3. The state of development of the instruments nowadays available for safeguards purposes was to be demonstrated with respect to their mode of operation and tamperproofness.
4. Practical experience was to be gathered to permit judgement of the effort necessary for exercises like this one and for the proposed safeguards system in general.
5. It was to be investigated, whether or not the operator's data could

be used to establish the material balance according to the methods outlined in [3].

6. The importance of the different strategic points [2] was to be calculated to establish priorities for the development of the different instruments and methods.

## 1.2. Safeguards Measures.

Three types of safeguards measures have been defined for the exercise. All measures, described in detail under 1.3. of this paper, have been established in such a way that, on the one hand, the material balance could be set up by only measuring the flow of plutonium at strategic points, i.e. without knowledge of the details of the production steps in the inner part (i.e. between the strategic points) of the plant. On the other hand, the measures had to be non-intrusive on plant operation as far as possible. Within these limits three types of measures have been set up as follows:

### i. Measures for the execution of the material balance.

In this category all the plutonium input and output streams were measured according to the principles of safeguarding nuclear material at strategic points. These measures are listed in Table I and are described in detail under 1.3. of this paper.

### ii. Measures used to establish a certain degree of containment.

Within this first exercise it was not possible to establish and maintain a complete containment as required in [1] (i.e. a tight containment). This was on account of the specific layout of the plant. Therefore only two containment measures were set up namely identification and sealing. The first one was performed to avoid or detect internal mixing of different types of material, which occurred for example after the final clean-out of the plant. The sealing measures were set up to avoid recycling of

the measured product, which in fact would be a very simple possibility to falsify the material balance and hence camouflage an internal diversion. The details of these containment measures are presented in Table II and described further under 1.3.

- iii. Table III contains the special measures. The reasons for the introduction of these special measures are due to the specific layout of the ALKEM plant. For example, it was not possible to measure the input stream within the Pu-storage but in the first box of the ceramic line. The transport of the material between Pu-storage and this first box had to be safeguarded by observation of the material transferred. This observation of the transport was a special measure. Another special measure was the clean out of the plant at the beginning of the campaign to establish well defined starting conditions. The details of these special measures are described under the following point.

### 1.3. Plant layout and the Identification of the Strategic Points

#### 1.3.1. Plant layout

The ALKEM fabrication plant is located on the premises of the Karlsruhe Nuclear Research Center; in this plant 200 kg of Pu are handled annually under commercial conditions. The layout of the plant is presented in Fig. 1 where it can be seen, that the plant is subdivided into different departments.

The functions of these departments are:

1. Plutonium storage.

The amount of Pu to be handled is stored here after arrival. This storage is used also for intermediate storage during operation and, thirdly, for final product storage before transportation.

2. Ceramic operations.

In the ceramic department the following steps of the operation are performed:

- a. Weighing and sampling of the material
- b. Calcination of the oxalate coming from the waste recovery and determination and regulation of the specific surface of the powder
- c. Mixing and homogenizing of  $\text{PuO}_2$  and  $\text{UO}_2$
- d. Pelletizing of the powder
- e. Sintering of the pressed pellets
- f. Breaking of defect pellets

### 3. Metallurgy

During the exercise this room was used only as the location for the calorimeter because this room is fully air conditioned.

### 4. Analytical Laboratory

The samples taken in the different departments are analyzed here for production control.

### 5. Waste analysis and health physics

All waste streams are collected in this department, measured by the neutron count method and separated into recoverable and non-recoverable wastes. This room is also the station of the health physics personnel.

### 6. Waste recovery

The amounts of recoverable waste separated in the waste analysis department are reprocessed in this department up to the oxalate step. The oxalate is transported to the ceramic department to be calcined. This department was not used for reprocessing of the material safeguarded.

### 7. Caisson 1

In caisson 1 the sintered pellets are ground, controlled and assembled into columns.

## 8. Caisson 2

In caisson 2 the columns are filled into the cans. The cans are welded and tested for leaks. The pins are transported to Pu-storage.

### 1.3.2. Identification of the Strategic Points

In Fig. 1 the strategic points or strategic areas are presented.

In identifying the strategic points two different sets of criteria had to be taken into account:

- i. According to the principles of the control of the flow of fissile material at strategic points [1] [5], all the input and output streams had to be measured.
- ii. The layout of the plant and the availability of the required instruments had to be taken into account. For example, the only hood, containing balances was the glove box K 01/85; hence, this box was identified as a strategic point.

In Table IV the strategic points are listed. Fig. 2 is a representation of the flow of fissile material through these strategic points. The description of the strategic points and the measures established there are given below.

#### 1.3.2.1. First Strategic Point: Storage of Plutonium and Box 1/85

The two parts of this strategic point are located in different sections of the plant. However, they have to be considered as one strategic point because the analysis and safing of the input streams and the scrap stream could be guaranteed only by using both parts. For future plants it can be assumed, that this splitting of the first strategic point will not be necessary [5], because a glove-box can be made to be available in the plutonium storage with the required instrumentation for input and scrap analysis.

At the ALKEM plant, the quantitative analysis of input and scrap streams can be carried out only in box 1/85. Therefore, additional containment measures and special measures had to be taken to guarantee that the input stream came into this box. These measures are presented in Table V.

#### 1.3.2.2. Second Strategic Point: Box CK 07/C1

The principles of the safeguards system require the measurement of the output streams. Normally and in future the calorimetric method and other indirect methods [5] being developed now will be used. At the conceptual design stage and at the beginning of the exercise it was not clear, whether or not the first prototype calorimeter which was constructed at that time, would give sufficiently good results. In order to be sure that the material balance could be closed in spite of a malfunction of the calorimeter, a strategic point was installed to measure the content of plutonium of the pellets. The box CK 07/C1 is the last point, where the plutonium is in a directly accessible form and therefore this point was selected. In the course of the exercise it turned out that the calorimeter worked reliable and with sufficient accuracy but the check of the accuracy of the calorimetric measurements could be carried out only with the results of the  $\gamma$ -spectroscopy of the measurement of the pellets in box CK 07/C1. The measures used at this strategic point are presented in Table VI.

#### 1.3.2.3. Third Strategic Point: Calorimeter

The calorimeter was installed in room 60, which is the only room fully air conditioned. The final product in form of pins were measured by the calorimetric method. For various reasons the results of the calorimetry were less accurate than the results of the  $\gamma$ -spectroscopy, as is shown under 6.4. However, the results of the calorimetry were able to indicate that between strategic points 2 and 3 material was neither withdrawn nor added. The measures used at this strategic point and the measures for safing the measured waste are presented in Table VIII.

#### 1.3.2.4 Fourth Strategic Point: Analysis of the Waste in Room 89

This strategic point also was fixed by the layout of the plant. A counting device for spontaneous and  $\alpha, n$  neutrons was used to provide the necessary analysis of the waste. The measures used at this strategic point and the measures for safing the measured waste are presented in Table VIII.

### 1.4 Description of the Production Campaign

The data of the production campaign safeguarded during the exercise are

given in Table IV. The order concerned the production of 200 kg of ceramic material, i.e. 200 kg of U-Pu mixed oxide fuel pins with a Pu concentration of 2.3 %.

## 2. The Course of the Exercise

### 2.1. Description of the instruments

Under this paragraph the instruments used during this exercise and their mode of operation are described.

#### 2.1.1. Input and Scrap Analysis

The instruments and methods for the input and scrap analysis at the first strategic point, box 1/85, were balances and chemical analyses. The first balance was a balance with a range of 4 kg and an accuracy of  $< 0.1$  g (type: Mettler K 4 T) and the second balance had a range of 100 g with an accuracy of  $< 1$  mg (type: Mettler 46 T). Before and during the exercise these two balances were gauged with high precision weights and no significant difference was detected. The methods of chemical analyses performed by the Institute of Radiochemistry were the AGO-method and the Coulorimetry. The results and a description of the statistical analysis of these methods are given in Chapter 6. Further details of the experiences gained at this strategic point are given under 2.2.

#### 2.1.2. Gamma-Spectroscopy

$\Gamma$ -spectroscopy was used to measure the amount of plutonium in the product (pellet) stream. Because of the large number of pellets produced (ca. 12000) a special sampling method was developed and used. More details of the statistical analysis are given in Chapter 6, in this paragraph, the apparatus as such and the mode of operation is described.

The  $\gamma$ -spectrometer is shown in Fig. 3. The  $\gamma$ -rays of one single pellet are counted with the use of a NaI crystal and registered by a multichannel device. The multichannel device contained 400 channels, with which the  $\gamma$ -ray spectrum was measured between 300 and 460 keV. In this range of the spectrum the peaks of Pu-239 (385 keV average) and U-237 (333 keV) were measured.

The concentration of U-237 on the one hand depends on the time elapsed between the last irradiation and the measurement. Due to the short half-life of U-237 of 6.75 d and due to the fact that more than 70 d have passed, i.e. more than 10 half-lives, this first dependence on the U-237 concentration could be neglected. On the other hand the equilibrium concentration of U-237 depends linearly on the Pu-241 concentration. As it was found by mass-spectrometrical analyses of the input stream, that the isotopic composition was homogeneous, the Pu-241 concentration in the same way as the Pu-239 concentration reflected the total amount of Pu in the respective pellet. Therefore, the pulses generated by the U-237 could be used as a relative measure of the Pu content of a pellet together with the pulses generated by the Pu-239. That means, the whole number of pulses between 300 and 460 could be used as that relative measure. It was assumed, firstly, that the number of these pulses was proportional to the amount of Pu in a pellet and, secondly, that the Pu was homogeneously distributed in the respective pellet. The last assumption could be tested and accepted by the statistical means described in 6.2. The first assumption is due to  $\sqrt{6}$ .

The measurement procedure was set up as follows:

- i. A random sample of 30 pellets was drawn from each batch of pellets, which contained ca. 600 pellets.
- ii. Each of these 30 pellets was measured in three different positions and one background measurement was taken between each pellet measurement.
- iii. One of these 30 pellets measured was separated as the standard pellet for chemical analysis.
- iv. This standard pellet was measured 30 times with one background measurement between each measurement.
- v. The standard pellet of the first batch was kept in the box and was measured prior to the measurements of each batch to detect possible shifts of the instrument.

The principle of the determination of the Pu-content of each batch was this: The combination of a relative measurement by  $\gamma$ -spectroscopy of randomly

sampled pellets with an absolute measurement by chemical analysis allowed a calculation of the Pu-content of each batch of pellets. The principles of the statistical analysis of these measurements are outlined in paragraph 6.3.

### 2.1.3. Calorimeter

The calorimetric method was used in the exercise to determine the amount of plutonium in the final product, the pins. The results of the calorimetry could be checked to a certain degree by testing these results against the results of the  $\gamma$ -spectroscopy. This testing procedure and the evaluation of the results of the calorimetry are given in Chapter 6; in this paragraph, only the instrument and the method of measurement are described.

The principles of calorimetry are well known and often applied to the measurements of heat fluxes. Calorimetry can be used also to measure the heat flux generated by the  $\alpha$ -decay of plutonium and Am-241 in Pu-containing fuel. For a known isotopic composition (incl. Am-241) of the fuel, the amount of Pu can be calculated from the heat flux generated by the different isotopes of Pu and by Am-241.

The principle of the first pin-calorimeter used during the exercise is shown in Fig. 4. The device consisted of two chambers, the measurement-chamber and the comparison-chamber, enclosed in a water bath with constant temperature. In these chambers, the heat flux induced by  $\alpha$ -decay of the Pu and Am-241 in the pins generates a potential difference in the thermocouples which surround the chambers and are connected in series. The potential difference can either be measured separately for each chamber by a micro-voltmeter or the potential differences of both chambers can be balanced in a Wheatstone-bridge. The measuring time is the time required for an equilibrium of the respective heat fluxes of the two chambers to be established.

A typical curve of such a measurement drawn by the recorder attached to the Wheatstone-bridge is shown in Fig. 5. The interesting figure is the difference in  $\overline{mV}$  between the heat flux of the measurement chamber and the comparison chamber; this measured value can either be taken from the record curve, or can be ascertained directly by measuring the voltage of each chamber.

Normally and in future applications a heating device will be installed in the comparison chamber. For this first application of this prototype

calorimeter three steps of operation were performed:

1. Relative measurement

One standard batch containing 10 long pins was selected and against this standard batch all the other batches were measured.

2. The standard batch was gauged by means of a heating device (electrical resistance) installed in the measurement chamber.
3. The two chambers were balanced against each other by heating devices in each chamber which were heated up with the same power input.

The statistical evaluation of the results of the measurements and the discussion of the sources of error and of the reliability of the instruments are given in paragraphs 6 and 4. Further details about the measurements are given in 2.2.

2.1.4. The Neutron Counter

The neutron counter was used during the exercise to determine the Pu-content of the waste stream. Two types of waste entered this strategic point, i.e. liquid waste from the chemical analyses and dry waste, e.g. contaminated paper.

These two types were measured by the neutron counter and separated into recoverable and non-recoverable waste. The instrument and the two steps of operation are described in this paragraph. The principles of the neutron counting device are shown in Fig. 6. The spontaneous and the  $\alpha, n$ -neutrons of the Pu-containing waste are moderated by the paraffin bed, which surrounds the  $\text{BF}_3$ -counting tubes. The neutrons caused  $n, \alpha$ -reactions with the boron in the  $\text{BF}_3$ -counting tubes and these  $\alpha$ -particles were counted. The FHT 4064 counter registered the pulses and printed the number of pulses in a given period of time.

At the beginning of the exercise the instrument was standardized first. This was done by measuring several batches of waste with a Pu-content, determined by weighing and chemical analysis  $\underline{[6.5]}$ .

The procedure of measurement was set up in this way: Each batch of waste was measured five times, and afterwards the background also was measured

five times. With the statistical procedure described in 6.5. the amounts of Pu in the respective batches of waste were calculated.

## 2.2. Course of the exercise and Measurements at the Different Strategic Points

The time schedule of the activities at the different strategic points is given in Fig. 7.

2.2.1. During the first phase of the exercise the fissionable material arrived at Karlsruhe and the personnel of the exercise was trained. The plutonium was sealed in the Pu-storage in the manner shown in Fig. 8.

2.2.2. The second phase of the exercise was the clean-out from the plant of the material processed before the exercise. This clean-out was performed to provide well defined starting conditions. The clean-out was checked by controlling the books of the ALKEM company. In retrospect, this phase II was redundant to a certain degree because it was possible to carry out the separation between material under safeguards and that not under safeguards with sufficient accuracy by mass-spectrometrical analysis due to the specific, unique isotopic composition of the safeguarded material. However, this information was not available at the beginning of the exercise and this is why this phase II was performed.

2.2.3. During phase III the following measures and activities were performed:

i. Analysis of the input stream at the first strategic point.

The amount of plutonium to be processed during the exercise was transported to box 1/85. Then the material was homogenized by mixing, was weighed, and the samples for the chemical analysis were drawn out, one sample for ALKEM and one sample for safeguards purposes. Unfortunately, unexpected difficulties arose during this sampling procedure and therefore the sampling was repeated in such a way that the total amount was divided into three batches from which one sample each was drawn. This was done to check both the chemical analyses and the sampling procedure. The second reason for this repetition was due to major shipper-receiver difference (ca. 50 g) found during

the first sampling; this shipper-receiver difference was discovered also in the second sampling. Afterwards the input material was transported back to the Pu storage. From there it was removed in accordance with the requirements of production.

- ii. Preparation of the standards for the neutron counter and calibration of the neutron counter.

The standards were prepared from broken pellets of the first sintered batch of the production. This preparation consisted of three steps:

1. Milling and homogenizing of the broken pellets.
2. Identification of the material as safeguarded material by mass-spectrometrical analysis and determination of the Pu-concentration by chemical analysis.
3. Weighing of four batches with increasing amounts of Pu and scattering of the material into paper and plastic bags.

With these standards the neutron counter was calibrated. The statistical procedure is outlined in 6.4.

After calibration the waste stream was analyzed in a more or less continuous activity up to the end of phase V.

- iii. Installation and test runs of the  $\gamma$ -spectrometer.

During phase III the  $\gamma$ -spectrometer was installed in box CK 07/C1 and several test runs were performed.

2.2.4. Phase IV of the exercise consisted of three main activities:

- i. The pellets produced were measured by  $\gamma$ -spectroscopy according to the procedure described in 2.1.
- ii. The waste stream was analyzed.
- iii. The calorimeter was tested and finished with the first pins produced.

During phase IV all production steps in the ceramic department and all stages of pin production were performed.

2.2.5. Phase V of the exercise, the last phase, consisted of four activities:

- i. The scrap produced during the production was homogenized and analyzed by weighing and chemical analysis.
- ii. The Pu-content of the pins was determined by calorimetry. Some difficulties arose in measuring the pins by calorimetry due to the fact that the calorimeter was a prototype device. For example, it was planned to measure each batch of 10 long pins or 33 short pins in the measurement chamber against the heating device in the comparison chamber. Unfortunately, the heating device did not work reliably as well as the measuring time was too long. Therefore, the measuring procedure was divided in three steps (see 2.1.):
  1. Relative measurement of all pins against a reference bundle.
  2. Gauging the reference bundle by measuring against the heating device.
  3. Balancing of the two chambers with one heating device in each chamber.

The reason why the measuring time played an important role at this point was that within only 2 1/2 weeks all pins had

to be measured because of the fixed term of delivery of pins to the customer.

- iii. The plant management was required to perform a final clean-out of the plant for inventory taking. A certain amount of dust was found and this dust was analyzed and identified in the first strategic point. It was found [see 4.1] that not the whole quantity could pertain to material safeguarded during the exercises; a certain amount must have been due to the production campaign processed before.
- iv. The final measurements of the waste stream, i.e. the waste coming from the chemical analyses were performed.

After the end of the practical part of the exercise results were evaluated by the methods shown in Chapter 3.

### 2.3. Methods used for Data Collection

The system for the collection of data measured at the different strategic points was set up according to two objectives:

1. All data of the measurements and all measures performed had to be registered to establish a permanent record of the results.
2. The way of data collection and especially the forms used were arranged so, that the forms could be transformed directly into punched cards as an input for the computer program. It was planned to carry out the whole material balance and the statistics of the time spent by the safeguard personnel by means of a special computer program. This was performed to avoid unnecessary errors in transferring the measured data to input forms and to minimize the effort of calculating the material balance.

Unfortunately, the effort required to write the computer program was relatively high and moreover the material balance was calculated by hand in addition to the calculations by the computer to provide an accurate check of all the results.

However, it could be shown that it is useful to calculate the material balance at strategic points with the aid of a computer. It became clear also, that the full advantage of such a computerized system will not be attained for small exercises but for larger ones or for the final safeguards system using the concept of strategic points.

In Fig. 8 and 9 two typical forms are presented, the first one for the input and scrap analysis [Fig. 8] and the second one for the  $\gamma$ -spectroscopy [Fig. 9].

### 3. Survey of the Statistical Analysis of the Measurements

#### 3.1. Introduction

The statistical methods used for the statistical analysis are based on the theory of estimation and the theory of testing statistical hypotheses both of which are well known in the field of mathematical statistics. To apply these theories a number of definitions and assumptions have to be made and the principle of the material balance has to be shown from a statistical point of view. The objective of the statistical analysis of the exercise as well as one objective of the whole exercise was to verify, in a practical application, the statements made in [1] from a more fundamental point of view.

The principle of the material balance carried out in the exercise was this: The throughput measurement was carried out by measuring and analyzing all the input and output streams. The inventory taking as required in [1] did not consist in a set of activities but in the definition of a time date. The operator stated at September 19 that the plant was empty, i.e. that the production had been finished. Therefore, the process inventory was considered to be zero as the result of inventory taking. This type of inventory taking can be assumed to be accurate, because any material still in the plant would be declared by the safeguards people as diverted (of course with a certain probability).

For statements about diversion according to the principles in [1], three models for the practical evaluation of the probability of diversion and of the risk of detection have been developed and considered.

The first of these models is based on the Bayes' approach to statistical inference. With this model the statements from [1] can be verified in a

complete manner. Unfortunately, this Bayesian method can be applied with advantage only, if two conditions are fulfilled:

- i. It must have been shown that the material remaining in a plant can be regarded as a random variable. This means, for example, that the material unaccounted for is a random variable.
- ii. The probability distribution (the prior probability distribution) of the random variable, i.e. the material remaining, must be known.

For the first exercise no data were available to test and meet these two conditions and so the Bayesian method could not be applied, but only the Bayesian model with vague prior information, which gives the same results as the method of confidence intervals.

The second model [8] is based on a purely testing procedure. The principle is that the data of the throughput measurements are evaluated in such a way that at least two independent sets of throughput measurements can be formed. With the first set of the throughput measurements a hypothesis is developed about the amount diverted and with the second set this hypothesis is tested. The main difference between the first and the second model lies in the fact, that by the second model the probability of diversion cannot be evaluated directly. This is on account of the reason, that the second model cannot use the prior information at all, i.e. in the way this can be used by the first model. However, some practical difficulties arose in applying this model to the exercise:

- i. The variances of the different instruments must be known, i.e. the data themselves cannot be used to test the measurement errors of the instruments.
- ii. For a small number of repetitions of each measurement, as was the case, the areas of acceptance of the non-diversion hypothesis would have been too large.

This means, that the second model could not be used for the evaluation of the statements, because firstly, no quantitative knowledge about the accuracies of the instruments was available a priori and, secondly, because some measurements could not be repeated. Hence, it would not have been possible to establish two independent sets of throughput measurements.

The third model uses an estimation procedure based on the theory of confidence intervals. Here, only the principles will be outlined. The result of a material balance consists of two quantities. Firstly, it consists of an estimate  $\bar{X}$  about the material unaccounted for, which may have been diverted or simply may have been forgotten by the operator inside a plant. Secondly, the result consists of the variance  $\sigma^2$  of  $\bar{X}$  or the estimate  $S^2$  of  $\sigma^2$ . The estimate  $S^2$  can be calculated in such a way, that if  $S^2$  is used for  $\sigma^2$ ,  $\sigma^2$  is larger than  $S^2$  only in  $1-\gamma$  % of the cases in the long run, which means

$$P(S^2 > \sigma^2) = 1 - \gamma$$

The model contains two steps:

1. Construction of a so-called confidence interval at a chosen level  $P$ .
2. The calculation of the risk.

The details of these two steps are described in 3.2.

The probability  $P$ , the confidence level, can be defined as probability of diversion, but only before the analysis. After construction of the confidence interval this probability  $P$  becomes a likelihood of diversion according to the usual definition of probability. The risk of diversion, which in principle is a probability statement prior to the decision about the statements on diversion, can be calculated with this model also. The calculus of the material balance including the statements is developed in this paragraph with the aid of this third model.

The calculation of the throughput measurement required a large number of additional statistical tests and other methods outlined in the appendix (6.) These additional tests were necessary because no or only a vague knowledge about the errors of the measurements by instruments was available before use of the instruments and analytical methods within the exercise.

### 3.2 Calculation of the Material Balance and of the Statements

3.2.1 After determination of the estimates of the true values of the respective input and output streams as well as the respective variances, as shown in the appendix, the estimate of the material balance can be

calculated very easily:

$$M = \sum_i \lambda_i M_{tot_i} \quad (3.2.1)$$

$$\sigma_M^2 = \sum_i \sigma_{M_{tot_i}}^2 \quad [i : \text{all input and output streams}]$$

$$\lambda_i = \begin{cases} +1 & \text{if } M_{tot_i} \text{ is the estimate of the input stream} \\ -1 & \text{elsewhere} \end{cases}$$

where

- $M$  = the estimate of the material balance or the estimate of the so-called MUF
- $\sigma_M^2$  = the variance of  $M$
- $M_{tot_i}$  = the results of  
input analysis  
scrap analysis  
waste analysis and  
either  $\gamma$ -spectroscopy or calorimetry
- $\sigma_{M_{tot_i}}$  = the variances for the respective  $M_{tot_i}$ .

The result obtained with the use of  $\gamma$ -spectroscopy has been calculated to:

$$M_1 = 48.38 \text{ gms}$$

$$\sigma_{M_1} = 8.09 \text{ gms}$$

and on the basis of the calorimetry

$$M_2 = 44.19 \text{ gms}$$

$$\sigma_{M_2} = 16.85 \text{ gms.}$$

Both the results are not significantly different, as can be seen immediately because both the estimates lie inside of the  $\pm 1-\sigma$  ranges of each other.

Due to the higher accuracy, the first value was taken to represent the result of the material balance to be used for calculations of the statements. The second result  $M_2$  was used as a confirmation that the first result is correct as well as no diversion occurred between the second and the third strategic point.

3.2.2 For the calculation of the statements established in 1 the following methods were used. The principal idea of this procedure is described in 3.1. For practical purposes, as in the case of the exercise, a method directly based on the theory of confidence intervals can be applied, because of the one-to-one correspondence between testing procedures and the construction of confidence intervals. By means of the method of confidence intervals the probability of diversion can be established as follows:

If one asserts that the interval  $I = (M - u(P) \cdot \sigma_M, M + U(P) \cdot \sigma_M)$  covers the real amount  $\mu$  diverted, I will include  $\mu$  with the probability  $P$ . This probability  $P$  represents the probability of diversion as established in 1, but only from an a priori point of view. With a fixed, i.e. measured quantity  $M$ , the diversion of at least  $I_u = M - U(P) \cdot \sigma_M$  can be stated only with the likelihood of diversion  $P$ . For this exercise, the lower boundary of the interval  $I$  was calculated for different likelihoods.

The quantity  $M$  was assumed to be distributed according to a normal distribution, firstly, on account of the knowledge of the normality of the different  $M_{tot}$ , and, secondly, the central limit theorem. Therefore, for fixed  $P$ , the lower boundary of the amount declared as diverted was calculated:

$$d_o = I_u = M - U(P_D) \cdot \sigma_M \quad (3.2.2)$$

with

$u(P)$  = the fractile of the normal distribution  $n(0,1)$  at the entry  $P$ .

Risk of Detection.

In contrast to the probability (likelihood) of diversion, the risk of detection represents a probability in the strict mathematical sense. The condition of evaluating the risk of detection is the knowledge of the amount diverted, because the risk represents a statement either of the operator or the system designer 1. As can be shown, the risk of detection (or the probability of detection as it is called in 8) can be calculated by:

$$R_D(d_o, P) = 1 - \phi\left(\frac{d_o + u(P) \cdot \sigma_M - \mu}{\sigma_M}\right) \quad (3.2.3)$$

where

$\phi (X)$  = the value of the normal distribution at the fractile X

$\mu$  = the actual amount diverted.

The results presented in 4.2. have been calculated with Equ. 3.2.2 and 3.2.3.

#### 4. Results of the Exercise

##### 4.1 Material Balance

The results of the material balance are shown in Table XVIII. The material balance was obtained by subtracting the estimates for the respective true values of the different output streams from that of the input-stream [ see 3.2 ]. The variances have been summed up, too, to obtain the variance of the difference between input and output, i.e. the material balance.

One point should be remarked in addition to Table XVIII, namely the amount of plutonium contained in that scrap material piled up during the final clean out. After homogenization of the material the amount of plutonium contained in this scrap material has been determined by chemical analysis to be 13 g. However, by mass-spectrometrical analysis, which was carried out to identify the material as belonging to the safeguarded campaign, it was detected that the isotopic composition differed significantly. The values found in that scrap are given in Table XIX. A comparison of the results with those of the campaign (PLATR) and those of the campaign processed before (SNEAK), proved the share of PLATR-plutonium in that scrap to be 6.5 g. The method used to calculate this value can be found in [ 10 ]. For the product stream, the results of  $\gamma$ -spectroscopy are listed together with the results of calorimetry. It is obvious that both results do not differ significantly, though the result of calorimetry has a much larger standard deviation due to the difficulties described in 6.4. which were caused by the prototype quality of the calorimeter used. Therefore, in this exercise, calorimetry was used mainly as a containment measure to assure, that no material could have been diverted between the pellet-sampling point and the final product.

#### 4.2. Statements about diversion

The statements about diversion are listed in Table XX. The probabilities (likelihoods) of diversion were calculated according to the principles mentioned in 3.2.

The actual amount of Pu withdrawn by ALKEM authorities during the exercise was 42 gms of plutonium. This actual amount withdrawn lies within the  $1-\sigma$  range of the material balance. On the other hand, it was possible to state (of course without any knowledge of the amount withdrawn) that with 95 % probability  $\geq 35.06$  gms of Pu had been diverted from the process stream.

The risk of the operator can be calculated by the straight-forward procedure shown in 3.2. The risks of the operator for different amounts are indicated in Table XX together with the corresponding probabilities of diversion. In the diversion of 42 gms the risk of the operator of detection was relatively high, or more precisely, in withdrawing 42 gms, the operator runs a relatively high risk of detection  $R_D$ , that a fairly large fraction of the amount withdrawn could be stated as diverted with a significant high probability of diversion. Hence, the operator did not take a significant low risk ( $< 10\%$ ). The ratio  $S = E/\sigma_E$ , which is a measure of the significance to a certain degree was found to be  $S = 6$  and therefore the fact of diversion could be detected beyond any doubts. For instance, the operator ran a risk of 21.8 %, that with probability (likelihood) 95 %  $> 35$  gms (i.e. 83 % of the amount withdrawn) could have been declared as diverted. This risk of 21.8 % is not significantly small and therefore, from a statistical point of view, the result (the fraction stated as having been diverted) cannot be called a rare event.

#### 4.3 Effort involved in the Exercise

In Table XXI, the efforts involved in the different safeguards activities are presented. These efforts are divided into three groups:

1. Man-hours spent for the safeguard activities inside the ALKEM plant

The effort expressed in man-hours is presented as a function of the strategic points. By far the largest fraction was spent on  $\gamma$ -spectroscopy, i.e. 53 %. The main reason was that two persons were necessary to carry out the  $\gamma$ -spectroscopy. In future, the effort spent

on  $\gamma$ -spectroscopy will no longer be required because the pins or subassemblies will be measured by calorimetry only.

## 2. Effort on chemical analyses

The number of chemical analyses according to the different streams is indicated in the Part b of Table XXI. The main fraction of the effort (66.7 %) was spent on analyses of the standard pellets of  $\gamma$ -spectroscopy also.

## 3. Effort on mass-spectroscopic analyses

In Part c of Table XIX the number of mass-spectroscopic analyses is listed. As mentioned in chapter 1, these analyses were required for

- i. Identification of material as being material under safeguards.
- ii. Determination of the value of the heat flux of the material for calorimetry.

For the latter purpose, the results of all analyses were taken into account. However, with known accuracy of the mass-spectrometer, at least five analyses mentioned in Table XXI would have been necessary.

During the first and the second phase of the exercise four members of the safeguards group were present at the strategic points. After the starting period, this number could be reduced to two persons throughout the months of August and September, when some of the main activities were performed.

### 4.4 Relative Importance of the Strategic Points

As shown in  $\sqrt{2}$ , the standard deviations of the estimates of true values for the different material streams can be regarded as an index of the importance of the respective strategic points. However, in establishing comparable quantities, the effort involved in obtaining the different standard deviations must be taken into account.

As an extension of [2] one can define the importance I:

$$I = \sqrt{A_i} \cdot \sigma_i \quad (4.4.2)$$

where

$A_i$  = the effort to obtain  $\sigma_i$

$\sigma_i$  = the standard deviation at the strategic point i.

The relative importance of the strategic points is defined as:

$$Z_i = I_i / \sum_{i=1}^m I_i \quad (m = \text{number of strategic points}) \quad (4.4.3)$$

As a first approximation the following figures were introduced to calculate A:

1 man-hour	=	10 cost units
1 chem.analysis	=	100 cost units
1 mass-spectr. analysis	=	250 cost units

The relative importance can be interpreted as follows:

It may be seen from Table XXII, too, that the index  $\sigma_i \sqrt{A_i}$  increases linearly with increasing standard deviation  $\sigma_i$  and increases by square root with increasing effort  $A_i$ . Since the objective of any safeguards system is to reduce the effort or the standard deviation, the absolute value of this index gives an idea on the work to be performed in improving the properties of the system or improving the so-called cost-effectiveness. This means, that this index indicates a measure of the improvement to be performed at the different strategic points. The higher the value of this index, the higher is the importance of the respective strategic point with respect to future improvement. The quantity Z can therefore be considered to give a measure of the relative importance of the various strategic points.

These fractions  $Z_i$  were calculated and are shown in Table XXII. According to the experiences made in this exercise, the improvement of either the accuracy of the calorimetry or the costs of the mass-spectrometrical analyses in relation to the other costs has the greatest influence on the improvement of the properties of the system considered in this exercise. Likewise it can be seen from Table XXII that the importance of the waste analysis was found to be relatively low.

## 5. Conclusions

In accordance with the objectives outlined in 1.1., the following conclusions can be drawn:

- i. The results of the exercise show the possibility of safeguarding a batch campaign exclusively at strategic points in an existing plant. The special measures performed were only necessary because of the specific layout of the ALKEM plant, in the future these measures will be redundant.
- ii. The material balance could be established with fairly small measuring accuracies; hence, the diversion of a small quantity could be detected with a high probability, i.e. about 80 % of the material withdrawn could be stated as having been diverted with a probability of 95 %.
- iii. The effort required for this first exercise was relatively large. However, this effort cannot be extrapolated to future exercises or the safeguards system as such because, firstly, the necessary manpower will be significantly ( $\approx 50\%$ ) reduced due to the absence of  $\gamma$ -spectrometry and, secondly, because more experience with the instruments will be available, i.e. the hypotheses about the accuracies of the instruments will be given.
- iv. The state of development of the instruments used can be regarded as being fairly sufficient for relatively small plants, but only sufficient as to their accuracies. As to the tamper-resistance the different activities in the framework of the SpFK project for the development of the instrumentation are absolutely necessary. From

the results, the relative importance, the priorities for these developments could be obtained, i.e. the highest priority has to be given to the calorimetry. A surprising result was the fact, that the analysis of the waste stream could be established to a satisfactory degree with a mean accuracy of 8 %. So that a high priority for the improvement of the accuracy of this stream will not be necessary.

- v. During the exercise, the ALKEM company spent approximately 150 man-hours on the exercise. On the other hand, the results of the waste analysis, on which 120 hours were spent by the safeguards personnel, could be used by the ALKEM people for their own material balance without further effort if any. One can state therefore, that no actual disturbance of the plant's operation took place during the exercise.

#### Acknowledgement

The authors want to thank Prof. Häfele, Dr. Gupta, Dr.A. von Baeckmann (IRCH) and Dr. Schneider (ALKEM) for valuable contributions to the exercise and for valuable discussions.

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## 6. Appendix: Statistical Analysis of the Measurements

### 6.1. Definitions and Assumptions

For the statistical analysis, a number of definitions and assumptions have been made:

1. The parameters of a distribution function or of the related density function are represented by Greek letters. These parameters are the true values (normally unknown) underlying a measurement. The mean value and the variance of the normal distribution are represented by  $\mu$  and  $\sigma^2$ , respectively. The normal distribution is referred to as  $n(\mu, \sigma^2)$ .
2. The estimates or statistics (calculated from a sample) of the parameters of a distribution are represented by Latin letters. The estimates for the parameters of  $n(\mu, \sigma^2)$  are represented by  $\bar{X}$  and  $S^2$ , respectively.
3. It is assumed throughout the whole statistical analysis that at least the mean values  $\bar{X}$  of a sample from the different measurements are distributed according to  $n(\mu, \sigma^2)$ . This assumption holds, on the one hand, due to the central limit theorem and, on the other hand, because of the asymptotic normality of maximum likelihood estimates, which are used to estimate the parameters of the respective measurements distribution. The assumption about normal variates has several consequences:
  - i. The mean value  $\bar{X}$  of a sample of size  $n$  of observations belonging to  $n(\mu, \sigma^2)$  is distributed according to  $n(\mu, \frac{\sigma^2}{n})$ .
  - ii. The ratio  $n \cdot S^2 / \sigma^2$ , where  $\sigma^2$  is the true variance and  $S^2$  the variance of a sample of size  $n$ , is distributed according to a  $\chi^2$  distribution with  $n-1$  degrees of freedom.
  - iii. The ratio  $S_1^2 / S_2^2$ , where  $S_1^2$  and  $S_2^2$  are variances of samples with sizes  $n_1$  and  $n_2$ , respectively, is distributed according to an F-distribution with  $n_1-1$  and  $n_2-1$  degrees of freedom.

4. In general, it is assumed, that different measurements are independent of each other. However, this assumption has been tested at different stages of the analysis. The measure by which independence can be tested, is a test for zero of the so called correlation coefficient. However, the acceptance of the hypothesis of a correlation coefficient having the value zero is only a necessary condition of independence but no sufficient condition is established in cases where normal distributions cannot be assumed for single measurement. As only the correlation coefficient is tested, a certain degree of assumption remains for the establishment of the independence of different measurements.
5. In testing hypotheses at the different steps of the statistical analysis the probability of the error of the first type was defined to be  $\leq 5\%$ . This is a usual value in applications of statistical methods. The formal procedure of a statistical test normally contains two steps, firstly the calculation of a critical region (whose limit is the confidence limit) for a specific hypothesis and secondly the rejection of the hypothesis, if the value to be tested belongs to the critical region. The alternative to the rejection of a hypothesis is defined as the acceptance of the hypothesis and this definition becomes more and more customary in the literature about statistics.
6. Normally, the term accuracy is not used in statistics to describe measurement errors either of the random or the systematic type. Therefore, this expression is not used in this paper either. An error of a measurement can be divided into a random error and a systematic error. The random error is named either standard deviation, which is the positive square root of the variance and is an absolute error, or coefficient of variation, which is the standard deviation divided by the corresponding mean value. The systematic error is called systematic error or bias.

## 6.2. Statistical Evaluation of the Results of the Chemical Analyses

Chemical analyses were required for determination of the Pu-contents at different stages of the exercise. These stages were in particular:

- i. Analyses of the input material with a Pu concentration of about 87 % and one batch with 69 %.
- ii. Analyses of the standards of  $\gamma$ -spectroscopy with a Pu concentration around 2 %.
- iii. Analyses of the standards of the neutron counter, in which scrap with a Pu concentration of about 2 % was scattered.
- iv. Analyses of the scrap material with a Pu concentration of about 2 %.

From prior experience of the chemical laboratory, the Institute for Radiochemistry, it was known, that it could not be assumed, that the standard deviations of the chemical analyses were the same for different Pu-concentrations. This hypothesis could be verified by the calculations. Therefore, the evaluation was made for the two different levels of concentration, i.e. 87 and 2 % respectively. The evaluation at each level of concentration consisted of five steps:

- 6.2.1. Test for equal variance of the different analyses.
- 6.2.2. Test for independence of the different analyses.
- 6.2.3. Analysis of Variance with all the analyses of the respective concentration level.
- 6.2.4. Test for acceptance of the hypotheses, if any, about the random error.
- 6.2.5. Calculation of the estimates of the mean values and of the combined variance for the different streams.

These procedures contain only the analysis of the random component of the analytical error. It was assumed that no bias had to be taken into account

because of the calibration of the chemical analyses by standards.

The five steps mentioned above will be outlined as an example of the evaluation of the input stream.

The input stream consisted of 4 batches. From each batch a sample was drawn out. Each sample was analysed, and each analysis consisted of at least 3 independent determinations. With the results of these analyses, as shown in Table X, the first steps were performed as described below. In Table X the values of the different sample means values  $\bar{X}_i$ , sample variances  $S_i^2$ , and coefficients of variation  $v_i$  were calculated.

#### 6.2.1. Tests for equal Variance were performed by F-tests.

The results of these tests are shown in Table XI. The test uses the fact, that the ratio of two sample variances is distributed according to an F-distribution, if the distributions from which the samples are drawn are normal ones with the same variance. In Table XI the critical values for the ratio of the sample variances  $S_i^2$  ( $i = 1,2,3,4$ ) and the actual values of these ratios are listed. As shown there, all the actual ratios are by far less than the critical ratios and, therefore, the hypothesis can be accepted at the 95 % level, that the variance of the four batches has the same value.

This value of the variance is as yet unknown, it is shown only that the same laboratory made the same random error in analyzing the four batches. The estimate of the variance is calculated in step 3.

#### 6.2.2. Test for Independence of the Four Batches

The test consists in testing the correlation coefficient  $\rho$  to be zero for all the 6 pairs of the four batches. To perform this test, the estimate  $r$  for the different batches had to be calculated by:

$$r_{ij} = \frac{\sum_{K=1}^{\min(n_i, n_j)} (X_{iK} - \bar{X}_i) (X_{jK} - \bar{X}_j)}{S_i \cdot S_j} \quad (6.2.1)$$

For a calculation of the estimates  $r_{ij}$  the sample sizes have to be equal. Since they are not equal for the input analyses, a subset of the larger samples was chosen by an additional random experiment, i.e. by random numbers.

For example to calculate  $r_{13}$ , from batch 1, two observations have been dropped by random numbers.

The statistic:

$$T_{ij} = r_{ij} \sqrt{\frac{\min(n_i, n_j) - 2}{1 - r_{ij}^2}} \quad (6.2.2)$$

follows a t-distribution with  $f = \min(n_i, n_j) - 2$  degrees of freedom for  $\rho = 0$ .

The results of the test are shown in Table XII. With these results the hypothesis can be accepted that the results of the analyses are uncorrelated, i.e. independent. The property of independence is necessary for step 6.2.5 to be carried out.

### 6.2.3. Analysis of Variance

In the third step, a so called Analysis of Variance was carried out. The objective was to calculate estimates of the mean values  $\mu_i$ , and of the variance  $\sigma^2$  of the four batches. It has been tested above that all the four batches have the same but unknown variance  $\sigma^2$ , which is the supposition of the method used here. It can be shown, that the estimates  $\bar{X}_i$  for  $\mu_i$  ( $i = 1, \dots, 4$ ) are given by:

$$\bar{X}_i = \frac{1}{n_i} \sum_{j=1}^{n_i} (X_{ij}) \quad (6.2.3)$$

and the estimate for  $\sigma^2$  by:

$$s_I^2 = \frac{1}{4(\bar{n}-1)} \sum_{i=1}^4 \sum_{j=1}^{n_i} (X_{ij} - \bar{X}_i)^2 \quad (6.2.4)$$

where

$$\bar{n} = \frac{1}{4} \sum_{i=1}^4 n_i$$

are the different sample sizes.

The results of the Analysis of Variance are listed in Table XIII, the estimate of the variance was found to be  $S_I^2 = 0.0320 \underline{\%}^2$ .

6.2.4. Before the exercise it was known that the coefficient of variation of the chemical analyses would be better than 0.2 % for a high concentration of plutonium.

Hence, this value of = 0.2 % has been taken as the hypothesis to be tested against the result of the Analysis of Variance. But each analysis  $i$  contains a number of  $n_i$  different observations and the result of the analysis of variance is the estimate of the variance of each element of the analyses. Therefore the variance of the analyses had to be divided by the number  $n_i$  of the observations and, therefore, the ratio:

$$q_i = \frac{4(\bar{n}-1)S_I^2}{n_i \cdot \sigma_i^2} \quad (6.2.5)$$

is distributed according to  $\chi^2$  with  $4(\bar{n}-1)$  degrees of freedom. The results of the test at the 95 % confidence level are shown in Table XIV. The result of the test shows that analyses of batches 1, 2 and 3 have a significantly lower coefficient of variation than 0.2 %, whereas, for batch 4, the hypothesis  $\delta = 0,2 \%$  could be accepted. To obtain the numeric values for the coefficient of variation shown to be necessary in step five, the upper limits of the 99.5 % confidence interval for the unknown variances were taken as these variances for calculation of the coefficients of variation. The upper limits  $\sigma_o^2$  are calculated by:

$$\sigma_o^2 = \frac{4(\bar{n}-1)S_I^2}{n_i \cdot \chi_{99.5}^2} \quad (6.2.6)$$

Therefore:

$$P(\sigma_i^2 < \sigma_o^2) = .995$$

The high probability was chosen, to have a sufficient likelihood that the true variances  $\sigma_i^2$  are less than or equal to the limit  $\sigma_o^2$  when the material balance is calculated. The corresponding coefficients of variation are listed in Table XIV also.

### 6.2.5. The Calculation of the Pu Content of the Input Stream

The amounts of Pu contained in the different batches of the input stream can be calculated as follows:

$$\mu_{\text{Pui}} = \mu_{\text{PuO}_{2i}} \cdot \bar{c}_i$$

where

$\mu_{\text{Pui}}$  = the amount of Pu in batch i

$\mu_{\text{PuO}_{2i}}$  = the amount of  $\text{PuO}_2$  in batch i

$\bar{c}_i$  = the concentration of Pu in  $\text{PuO}_2$  for batch i

The equation above holds also for estimates calculated from measurements:

$$\bar{m}_{\text{Pui}} = \bar{m}_{\text{PuO}_{2i}} \cdot \bar{X}_i \quad (6.2.6)$$

However, in using the estimates  $\bar{m}_{\text{Pui}}$ ,  $\bar{m}_{\text{PuO}_{2i}}$  and  $\bar{X}_i$  for the respective "true" values, the errors (i.e. random errors) have to be taken into account. It can be shown that the coefficients of variation  $\delta_{m_i}^-$  pertaining to respective estimates are related as follows:

$$\delta_{m_{\text{Pui}}}^2 = \delta_{m_{\text{PuO}_{2i}}}^2 + \delta_{X_i}^2 + \delta_{m_{\text{PuO}_{2i}}}^2 \cdot \delta_{X_i}^2 \quad (6.2.7)$$

This equation holds for any distribution of independent random variables, i.e. the relationship holds if weighing and chemical analyses are independent, which can be assumed with sufficiently good reasons.

For the exercise the weighing error  $\delta_{m_{\text{PuO}_{2i}}}^2$  was neglected, because it was found to be two orders of magnitude less than  $\delta_{X_i}^2$ . The amount of Pu in the input stream was calculated then by summing up the amounts of the four batches. The standard deviation was calculated to be the square root of the sum of the variances of the four batches. The results was found to be:

$$\bar{M}_{\text{tot input}} = 5070.25 \text{ g} \quad \text{and}$$

$$\sigma_{\text{tot input}} = 5.86 \text{ g}$$

The same procedure as described above had to be repeated for the Pu at the 2 % concentration level, which consisted of the following materials:

- i. Scrap
- ii. Standard pellets for  $\gamma$ -spectroscopy
- iii. Standards for the neutron counter

The final results of these calculations are given in chapter 4.

### 6.3. Analysis of $\Gamma$ -Spectroscopy

The determination of the Pu-content with the help of  $\gamma$ -spectroscopy uses the following simple relationship:

$$\mu_{\text{Pui}} = \Gamma_{\text{Li}} \frac{\mu_{\text{is}}}{\gamma_{\text{is}}} \quad (6.3.1)$$

where

$\mu_{\text{Pui}}$  = the amount of plutonium in a batch i of pellets

$\Gamma_{\text{Li}}$  = the number of  $\gamma$ -pulses of batch i

$\mu_{\text{is}}$  = the amount of plutonium contained in the standard pellet

$\gamma_{\text{is}}$  = the number of pulses of the standard pellet

Equation (6.3.1) can be written immediately for the estimates of the respective true values:

$$m_{\text{Pui}} = G_{\text{Li}} \frac{m_{\text{is}}}{g_{\text{is}}} \quad (6.3.2)$$

Since these estimates are random variables, the coefficient of variation  $\delta_{\text{Pui}}$  can be found to be with good approximation:

$$\delta_{\text{Pui}}^2 = \delta_{\text{Li}}^2 + \delta_{m_{\text{is}}}^2 + \delta_{g_{\text{is}}}^2 \quad (6.3.3)$$

The amount of plutonium in each batch  $i$  and the respective measurement error can be calculated by the equations 3.3.2 and 3.3.3. This requires several steps:

- 6.3.1. Determination of  $G_{\text{Li}}$  and  $\delta_{\text{Li}}^2$  by  $\gamma$ -spectroscopy from a sample of different batches  $i$ .
- 6.3.2. Determination of  $g_{\text{is}}$ , the number of  $\gamma$ -pulses of the standard pellet of the batch  $i$ .
- 6.3.3. Weighing, chemical analysis, and statistical analysis for determination of the Pu-content of the standard pellet.

The three steps are described below:

#### 6.3.1 Determination of $G_{\text{Li}}$ and $\delta_{\text{Li}}^2$

The method of measurement had been set up as follows: In each batch  $i$ , of about 600 pellets the pellets were counted. The result was  $m_i$ . Secondly a random sample of 30 pellets was drawn out to be measured. The number of 30 pellets was chosen for considerations of:

- i. The time available for measuring purposes without disturbing the fabrication process;
- ii. the accuracy, needed to achieve a sufficiently small combined variance of the whole  $\gamma$ -spectroscopy.

After removal of the 30 pellets each pellet was measured in three different positions. This procedure was to provide a possibility to test the homogeneity

of the plutonium inside the pellets. If the plutonium had been distributed inhomogeneously, different rates of  $\gamma$ -pulses would have been measured in different positions due to self-shielding effects. The procedure of evaluation of  $G_{Li}$  and  $\sigma_i^2$  was this:

- i. For each column  $j$  ( $j = 1, 2, 3$ ) of batch  $i$  the sample mean and the sample variance were calculated as

$$\bar{X}_{ij} = \frac{1}{30} \sum_{K=1}^{30} X_{ijK}$$

as the sample mean and:

$$S_{ij}^2 = \frac{1}{30} \sum_{K=1}^{30} X_{ijK}^2 - \bar{X}_{ij}^2 \quad (6.3.4)$$

$$(j = 1, 2, 3)$$

$$(i = 1, 2 \dots 26 \text{ batches})$$

- ii. The values  $\bar{X}_{ij}$  and  $S_{ij}^2$  were tested for equality with the following test:

If

$$\frac{(\bar{X}_{iK} - \bar{X}_{i\ell})}{S_d} > t_{1-(\alpha/2), f} \quad (6.3.5)$$

$$(K, \ell = 1, 2, 3 \ K \neq \ell)$$

the hypothesis of equal  $\bar{X}_{ij}$  ( $i$ -fixed) would have been rejected.

Where

$$S_d^2 = \frac{1}{30} (S_{iK}^2 + S_{i\ell}^2) \quad (6.3.6)$$

and

$t_{1-(\alpha/2), f}$  = the value of the t-distribution at the 97.5 % level with  $f = 29$  degrees of freedom.

This means that three tests had to be carried out for each batch i:

a)  $\bar{X}_{i1} : \bar{X}_{i2}$

b)  $\bar{X}_{i2} : \bar{X}_{i3}$

c)  $\bar{X}_{i1} : \bar{X}_{i3}$

Fortunately, no rejection occurred for each of the 78 tests (26 batches) and, therefore, the plutonium could be assumed to be distributed homogeneously inside the pellets.

iii. The variances  $\sigma_{ij}^2$  for  $\bar{X}_{ij}$  were estimated by taking the upper 95 % limit of the confidence interval constructed with the help of the  $S_{ij}^2$  and the  $\chi^2$  distribution with  $f = 29$  degrees of freedom at the lower limit, hence

$$\sigma_{ij}^2 = \frac{S_{ij}^2}{\chi_{0.05}^2(29)} \quad (6.3.7)$$

The factor  $n = 30$  in the numerator has disappeared because the variance of the mean value of a sample of  $n = 30$  is only  $1/n$  of the variance of a single element.

The values of  $\sigma_{ij}^2$  were calculated at the upper limit of the confidence interval so as to provide a likelihood of at least 95 %, that the true, but always unknown, values of the variances  $\sigma_{ij}^2$  are less than this limit.

iv. The estimate of the mean value and the variance of each batch is calculated then by combining the  $\bar{X}_{ij}$ 's and  $\sigma_{ij}^2$  according to the maximum likelihood principle:

$$\bar{X}_i = \frac{\sum_{j=1}^3 \bar{X}_{ij} / \sigma_{ij}^2}{\sigma_i^2} \quad (6.3.8)$$

where

$$\sigma_i^2 = \frac{1}{3 \sum_{j=1}^3 \frac{1}{\sigma_{ij}^2}} \quad (6.3.9)$$

These quantities  $\bar{X}_i$  and  $\sigma_i^2$  are the "best" estimates for the unknown mean values and variances, respectively, but without the background, which has to be taken into account in the next stage.

- v. Between each pellet, the background was measured. The measurements of the background therefore also contained  $n = 30$  values. As the next step, the independence between background and measurement has to be tested by the same procedure as shown in 6.2.2. Or more precisely, it has to be tested that the coefficient of correlation  $\rho$  cannot be regarded to be positive. Then the estimate of the mean value  $\bar{U}_i$  and the variance  $\sigma_{U_i}^2$  of the background had to be calculated by Equ. 6.3.4 and 6.3.7.

- vi. With all these quantities, the values of  $G_{Li}$  and  $\delta_{Li}^2$  could be calculated:

$$G_{Li} = m_i (\bar{X}_i - \bar{U}_i) \quad (6.3.10)$$

where  $m_i$  is the number of pellets in batch  $i$ .

And:

$$\sigma_{GLi}^2 = m_i^2 (\sigma_i^2 + \sigma_{U_i}^2) \quad (6.3.11)$$

and therefore:

$$\delta_{GLi}^2 = \frac{m_i^2 (\sigma_i^2 + \sigma_{U_i}^2)}{m_i^2 (\bar{X}_i - \bar{U}_i)^2} = \frac{\sigma_i^2 + \sigma_{U_i}^2}{(\bar{X}_i - \bar{U}_i)^2} \quad (6.3.12)$$

### 6.3.2. Determination of $g_{is}$ and $\delta_{gis}^2$

The determination of  $g_{is}$  and  $\delta_{gis}^2$  requires but a few steps. The standard

pellet of each batch has been separated, it has been measured 30 times in different positions and between each measurement the background has been measured.

The first step of the evaluation of  $g_{is}$  (the number of pulses of the standard pellet) and  $\delta_{gis}^2$  consisted in a test between background and measurement for independence, i.e. that there is no significant positive correlation coefficient. The procedure is the same as in 6.2.2. and 6.3.1. The second step consists of the calculation of

$$\bar{X}_{Si}, \sigma_{Si}^2, \bar{U}_{Si} \text{ and } \sigma_{USi}^2 \text{ with the}$$

aid of Equ. 6.3.4 and 6.3.7 where

$\bar{X}_{Si}$  = meanvalue of the 30 measurements of the standard pellet

$\sigma_{Si}^2$  = the variance of  $\bar{X}_{Si}$

$\bar{U}_{Si}$  = mean value of the 30 measurements of the background of the standard pellet

$\sigma_{USi}^2$  = variance of  $\bar{U}_{Si}$

With these quantities, the values of  $g_{is}$  and  $\delta_{gis}^2$  can be determined as

$$g_{is} = \bar{X}_{Si} - \bar{U}_{Si} \quad (6.3.13)$$

$$\delta_{gis}^2 = \frac{\sigma_{Si}^2 + \sigma_{USi}^2}{(\bar{X}_{Si} - \bar{U}_{Si})^2} \quad (6.3.14)$$

### 6.3.3 Determination of $m_{is}$ and $\delta_{gis}^2$

The variable  $m_{is}$  was defined as the estimate of the Pu-content of the standard pellet and  $\delta_{gis}^2$  the coefficient of variation of  $m_{is}$ . The determination of these two quantities was performed by chemical analysis and weighing.

The statistical analysis consists of all the steps described in 6.2 where

the procedure is shown for the  $n = 4$  input batches. The procedure for the standard pellets of the different batches is quite the same with the only exception that  $n = 26$  batches had to be considered. Therefore, the quantities  $m_{is}$  and  $\delta_{gis}^2$  could be calculated by this procedure.

#### 6.3.4. Calculation of the Plutonium Content of the Output Stream

The plutonium content was calculated for the output stream ( $\gamma$ -spectroscopy) with the help of the equations given below. Besides to the calculation several tests for independence (i.e. tests for  $\rho$  to be not positive) have been carried out between the measurements of different batches. The Pu-content can be calculated by:

$$M_{\text{tot output}} = \sum_{i=1}^{26} m_{\text{Pui}} \quad (6.3.15)$$

where the  $m_{\text{Pui}}$  are calculated with Equ. (6.3.2) and

$$\sigma_{M_{\text{tot}}}^2 = \sum_{i=1}^{26} m_{\text{Pui}}^2 \cdot \delta_{\text{Pui}}^2 \quad (6.3.16)$$

where the  $\delta_{\text{Pu}}^2$  are calculated with Equ. (6.3.3) and where

$M_{\text{tot output}}$  = Pu-content of the output stream

$\sigma_{M_{\text{tot}}}^2$  = variance of  $M_{\text{tot output}}$

The results of these calculations were found to be:

$$M_{\text{tot output}} = 4209.6 \text{ [g]} \text{ and } \sigma_{M_{\text{tot}}} = 4.9 \text{ [g]} .$$

#### 6.4. The Analysis of the Calorimetry

At the end of the exercise the calorimetric method could be used for the determination of the Pu-content of the pins produced. The principal

relationship for the calorimetry can be stated as follows:

$$\mu_{\text{Pu}} = \Omega_{\text{A}} / \omega_{\text{S}} \quad (6.4.1)$$

where

$$\Omega_{\text{A}} \text{ [Watt]} = \text{heat flux of the whole material}$$

$$\omega_{\text{S}} \text{ [Watt/g]} = \text{specific heat flux per gramm of chemical plutonium}$$

$$\mu_{\text{Pu}} \text{ [g]} = \text{amount of Pu inside the material investigated}$$

The measurable estimates of the different batches  $i$  belonging to the quantities of Equ. (6.4.1) are:

$$m_{\text{Pu}_i} = W_{\text{A}_i} / W_{\text{S}_i}$$

The estimated are random variables and their coefficients of variation are connected (approximation):

$$\delta_{m_i}^2 = \delta_{\text{A}_i}^2 + \delta_{\text{S}_i}^2 \quad (6.4.2)$$

if the measurements are independent. This can be assumed because the methods for determination of  $W_{\text{A}}$  and  $W_{\text{S}}$  are completely different.  $W_{\text{A}}$  is determined by calorimetry,  $W_{\text{S}}$  by mass-spectrometrical analysis. An additional source of error can influence the accuracy of the calorimetry and the respective error cannot be expressed in Equ. (6.4.2). This error is the error of inhomogeneity of the isotopic composition of the input material. Therefore, a test for the homogeneity of the isotopic composition had to be carried out, which is described in 6.4.1.

The evaluation of  $m_{\text{Pu}}$  was performed in two steps:

6.4.1. Determination of  $W_{\text{S}}$  and  $\delta_{\text{S}}^2$ .

6.4.2. Determination of  $W_{\text{A}}$  and  $\delta_{\text{A}}^2$ .

6.4.1 Determination of  $W_S$  and  $\delta_S^2$ 

The first step of the calculation of  $W_S$  and  $\delta_S^2$  consisted in a test of the homogeneity of the isotopic composition of the input material. From the 6 cans of the input material one sample each was taken for mass-spectrometrical analysis and was analyzed. The results of these six analyses were grouped into two groups by a random experiment with 3 analyses in each group. For these two groups, the sample means and sample variances were calculated for each isotope. The following test has been carried out (t-test):

If

$$t'_j = \frac{(\bar{x}_{1j} - \bar{x}_{2j})}{s_j} \sqrt{\frac{3}{2}} > t_{97.5,4} \quad (6.4.3)$$

then the hypothesis about homogeneity would have been rejected.

Where

$$s_j^2 = \frac{2 s_{1j}^2 + 2 s_{2j}^2}{4} = \frac{s_{1j}^2 + s_{2j}^2}{2}$$

$\bar{x}_{1j}$  = sample mean of the first group for isotope j

$\bar{x}_{2j}$  = sample mean of the second group for isotope j

$t_{97.5,4}$  = The value of the t-distribution at the 97.5 level with 4 degrees of freedom.

$s_{1j}^2$  = sample variance of the first group for isotope j

$s_{2j}^2$  = sample variance of the second group for isotope j

In the Tables the value of the t-distribution can be found as:

$$t_{97.5,4} = 2.776.$$

The highest value of all the tests was found to be

$$t'_{\max} = 0.474.$$

Therefore, homogeneity of the isotopic composition could be assumed.

Hence, the quantities  $W_{Si}$  were taken  $W_{Si} = \text{const } (i)$  and  $\delta_{Si} = \delta_S^2 = \text{const } (i)$ .

Another reason mentioned in 1.2. for this sampling of the input material was to obtain values for the different identification steps carried out with the different product streams.

After these tests for homogeneity the quantities  $W_{Si} = W_S$  and  $\delta_{Si} = \delta_S$  could be determined with the help of the following relations:

$$W_S \overline{[W/g]} = \sum_{i=238}^{242} k_i \bar{C}_i + k_{Am} \bar{C}_{Am} \quad (6.4.5)$$

and

$$\delta_S = \sigma_S / W_S \quad (6.4.6)$$

and

$$\sigma_S^2 = \sum_{i=238}^{242} \sigma_{C_i}^2 + \sigma_{Am}^2 \quad (6.4.7)$$

where

$k_i$  = specific heat flux per gram of the i'th isotope; these constants are known more or less precisely  $\overline{[10]}$

$\bar{C}_i$  = concentration of the i'th isotope in the material

Am = Americium 241

and

$\sigma_{C_i}^2, \sigma_{Am}^2$  = the variances belonging to the determined concentrations  $\bar{C}_i, \bar{C}_{Am}$ .

As on the one hand, no hypotheses about the  $\sigma_i^2$ 's, were available and on the other hand a large number ( $\leq 34$ ) of mass-spectrometrical analyses were carried out, the upper 95 % limit for the  $\sigma_i^2$ 's were calculated to be the values for  $\sigma_i^2$ . Equations (6.3.4) and (6.3.7) had to be adapted for that purpose:

$$\bar{C}_i = \frac{1}{n_i} \sum_{k=1}^{n_i} C_{iK}$$

and

$$\sigma_i^2 = \frac{S_i^2}{\chi_{0.05}^2(n_i-1)} \quad (i = 238, 242 \text{ Am})$$

with

$$S_i^2 = \frac{1}{n_i} \sum_{k=1}^{n_i} C_{iK}^2 - \bar{C}_i^2 \quad (6.4.8)$$

The values of  $n_i$  and the value of  $W_S$  and  $\delta_S$ , calculated with the help of equ. (6.4.5) to (6.4.8) are shown in Table XV. The values for  $W_S$  and  $S$  have been found to be:

$$W_S = 4.87644 \cdot 10^{-3} \quad \overline{[W/g]}$$

and

$$\delta_S = 0.45 \%$$

#### 6.4.2. Determination of $W_{A_i}$ and $\delta_{A_i}$

The procedures of the measurements in the determination of  $W_{A_i}$  and  $\delta_{A_i}$  will differ slightly between the first exercise and future exercises or future applications of calorimetry. Therefore, only a brief description of the statistical procedure used for the evaluation of  $W_{A_i}$  and  $\delta_{A_i}$  will be given here.

The procedure of the measurements within the exercise comprised these steps.

6.4.2.1 All the pins produced were classified in the following groups:

- a) 11 batches with 10 long pins, respectively
- b) 2 batches with 33 short pins, respectively
- c) 1 batch with 6 short pins
- d) 1 batch with 4 long pins and 1 short pin
- e) 1 batch with 4 long pins and 2 short pins.

6.4.2.2 The pins contained in the batches under a) and b) were measured against a reference batch, i.e. the first batch of a) has been regarded as a reference batch and the other 10 batches were measured once against that reference batch. The result was a set of differences  $\Delta V_i$  [ $\overline{\text{mV}}$ ], namely the difference of the voltage of the two chambers. The first and the second batch were measured 6 times in the process to provide an estimate of the variance of  $\Delta V_i$  and in order to detect possible shifts of the instruments.

6.4.2.3 The relationship between the voltage  $V$  and the heat flux  $W$  was established by measuring the voltage differences between the two chambers, the first chamber containing the reference batch, the second one containing the heating device. Different power inputs  $w_j$  were fed to the heat source and the related difference in the voltage  $v_j$  was measured.

6.4.2.4 The two chambers were balanced against each other. For this purpose, a heating source has been installed in each of the two chambers. Six different power inputs  $w_\kappa$  were fed to the two chambers (i.e. the same  $w_\kappa$  in both the chambers at the same time) and the voltage differences  $v_{\kappa\ell}$  were measured, where  $\ell$  is the number of repetitions of that balance experiment at the same power level  $w_\kappa$ .

6.4.2.5 The batches under points 1c) to 1e) were measured directly against the heat source at two different power levels.

The statistical analysis of these measurements was performed in the following steps.

6.4.2.6 The principal relationships for the batches measured relative to

the reference batch are:

$$W_{A_i} = W_{A_r} + \Delta W_{A_i} \quad (6.4.9)$$

where

$W_{A_r}$  = the heat flux of the reference batch

$\Delta W_{A_i}$  = the difference between the heat flux of the batch i and that of the reference batch.

The variances of  $W_{A_i}$  can be calculated by:

$$\sigma_{W_{A_i}}^2 = \sigma_{W_{A_r}}^2 + \sigma_{\Delta W_{A_i}}^2 \quad (6.4.10)$$

6.4.2.7 To determine  $W_{Ar}$  and  $\sigma_{W_{Ar}}^2$ , two different regression analyses had to be carried out:

- i. The first regression analysis with the values  $w_i$  and  $v_j$  from the measurements described under 6.4.2.3.

The result was a linear relationship between the power  $W_r$  and the voltage  $V_r$  for the reference batch:

$$W_{R_1} = \bar{a}_1 \cdot V_{R_1} + \bar{a}_2 \quad (6.4.11)$$

where, for  $\bar{a}_1$  and  $\bar{a}_2$  the respective variances (i.e. estimates of the variances)  $\sigma_{\bar{a}_1}^2$  and  $\sigma_{\bar{a}_2}^2$  could be calculated.

- ii. The second regression analysis with the values  $w_k$  and  $v_k$  from the measurements described under 6.4.2.4 was performed to obtain the line of equilibrium of the two chambers.

The result was:

$$V_{R_2} = \bar{b}_1 W_{R_2} + \bar{b}_2 \quad (6.4.12)$$

and in addition the estimate of the variance  $\sigma_{VR_2}^2$ .

iii. The point of intersection ( $V_{R_1} = V_{R_2}$ ) of these two regression lines represented the value  $W_{AR}$ , the heat flux of the reference batch:

$$W_{AR} = \frac{\bar{a}_1 \bar{b}_2 + \bar{a}_2}{1 - \bar{a}_1 \bar{b}_1} \quad (6.4.13)$$

were the quantities  $\bar{a}_1$ ,  $\bar{b}_1$ ,  $\bar{a}_2$ ,  $\bar{b}_2$  were determined by the respective regression analyses.

The value of the variance of  $\sigma_{W_{AR}}^2$  can be calculated by the following equation:

$$\sigma_{W_{AR}}^2 = (\bar{a}_1 K_1)^2 \left[ \frac{\sigma_{a_1}^2}{\bar{a}_1^2} + \frac{\sigma_{VR_2}^2}{K_1^2} \right] + \sigma_{a_2}^2 \quad (6.4.14)$$

and

$$\delta_{W_{AR}} = \left\{ \frac{(\bar{a}_1 K_1)^2}{K_1^2} \left[ \frac{\sigma_{a_1}^2}{\bar{a}_1^2} + \frac{\sigma_{VR_2}^2}{K_1^2} \right] + \frac{\sigma_{a_2}^2}{K_1^2} \right\}^{1/2} \quad (6.4.15)$$

with

$$K_1 = \frac{\bar{b}_1 \bar{a}_2 + \bar{b}_2}{1 - \bar{b}_1 \bar{a}_1} ; \quad K_2 = \frac{\bar{a}_1 \bar{b}_2 + \bar{a}_2}{1 - \bar{a}_1 \bar{b}_1}$$

6.4.2.8 From the first regression line, the values for  $W_{A_i}$  and  $\sigma_{\Delta W_{A_i}}^2$  established in Equ. (6.4.9) and (6.4.10) can be calculated to:

$$\Delta W_{A_i} = \bar{a}_1 \Delta V_i \quad (6.4.16)$$

$$\sigma_{\Delta W_{A_i}}^2 = (\bar{a}_1 \Delta V_i)^2 \left( \frac{\sigma_{a_1}^2}{\bar{a}_1^2} + \delta_{V_i}^2 \right) \quad (6.4.17)$$

The quantity  $\delta V_i^2$  was calculated from the variance between the repeated measurements of the reference batch against batch 2 (6.4.2.2).

With the aid of Equ. (6.4.9) to (6.4.16) all the values and the variances of the heat fluxes of the different batches could be determined. The coefficient of variation can be determined by, at least, as:

$$d_{W_{A_i}} = \sigma_{W_{A_i}} / (W_{A_i}) \quad (6.4.18)$$

6.4.2.9 To determine the values of the heat flux of the batches under 6.4.2.1c to 6.4.2.1e the procedure applied to the reference batch had to be repeated. This means that, for each of these batches the first regression line had to be calculated and then the point of intersection with the line of equilibrium had to be determined. The procedure was exactly the same as before in the determination of the reference batch.

In Table XVI some typical values of the errors calculated were listed. It can be seen there that neither the contributions of  $\Delta V_i$  nor the contributions of the line of equilibrium (l.o.e.) to the errors can be neglected. In the future, i.e. for the next application of calorimetry, both the contribution of  $\Delta V_i$  and that of the o.e. will disappear, because, firstly all batches will be measured against the heat source directly and, secondly, the instrument will be balanced to zero equilibrium.

### 6.4.3 Concluding Remarks on Calorimetry

With the aid of Equ. (6.4.2) the total amount of plutonium contained in the output (pin) stream can be calculated as:

$$M_{Pup} = \sum_{i=1}^k m_{Pui}$$

and

$$\sigma_{Pup}^2 = \sum_{i=1}^k \sigma_{m_{Pui}}^2 \quad (6.4.19)$$

The values of the Pu content have been calculated to:

$$M_{\text{Pup}} = 4213.76 \text{ [g]}$$

$$\sigma_{\text{Pup}} = 14.5 \text{ [g]}$$

The coefficients of variation of the different batches measured varied between:

$$\delta_{\text{min}} = 0.48 \%$$

$$\delta_{\text{max}} = 1.02 \%$$

The contribution to the coefficient of variation due to the determination of the isotopic composition was  $\delta = 0.45 \%$  as mentioned under 6.4.1. Hence, it is evident, that the error of the calorimetry as such is of the order of 0.1 % if all the batches are measured against the heat source. This is the next way of applying the calorimetry. However, these considerations show also above all, that the accuracy of determination of the isotopic composition must be improved in order to increase the accuracy of the whole method. This improvement lies mainly in the improvement of the determination of Pu-238, the error of which was found to be 1.3 %.

## 6.5 Analysis of the Waste Stream

The determination of the plutonium contained in the waste stream was performed with the aid of the neutron-counter, as mentioned in 2.3. The procedure comprised two steps.

### 6.5.1 Standardization of the instrument

### 6.5.2 Measurements

The two steps are described below.

#### 6.5.1 Standardization of the neutron counter

The standardization consisted of the following steps:

6.5.1.1. Four standards were prepared. For that purpose firstly appr. 360 gms of sintered scrap were homogenized, weighed, and analyzed by chemical analysis for determination of the Pu-concentration of the material. After that, the scrap was weighed in four batches of different quantities of material. The material of each of these batches was scattered carefully in paper and plastic material for the standards.

6.5.1.2. Each of these standards and several combinations of the standards were measured five times with an interval of one minute. In addition the background was measured.

The statistical analysis was performed in this way:

6.5.1.3. From the chemical analysis, the values of the Pu-concentration  $\bar{C}$  and of the coefficient of variation  $\delta_{\bar{C}}$  could be determined by Analysis of Variance. Since the material used had the same concentration level, the Analysis of Variance could be carried out in connection with the analysis of the standard pellets of  $\gamma$ -spectroscopy and the other scrap streams in the same way as described in 6.2. The results were:

$$\bar{C} = 2.038 \% \text{ Pu}$$

and

$$\delta_{\bar{C}} = 0.47 \%$$

The weighing error could be neglected and so a direct relationship could be established between the impulses measured and the Pu-content.

6.5.1.4. To establish the relationship between pulses and material, the following steps were performed:

- i. For each of the combinations  $i$  of the batches the sample means  $\bar{X}_i$  were calculated:

$$\bar{X}_i = \frac{1}{5} \sum_{K=1}^5 X_{ik} - \frac{1}{5} \sum_{K=1}^5 U_{ik} \quad (6.5.1)$$

where

$X_{ik}$  = the number of pulses during the time interval

$U_{ik}$  = background for the combination  $i$ .

Afterwards a straight line has been considered through the points calculated by [6.5.1] by the "least squares" method and the linearity was tested.

The straight line had the form:

$$S = \bar{a} \bar{X} + \bar{b} \quad (6.5.2)$$

where

$$\begin{aligned} S &= \text{amount of scrap in gms} \\ \bar{X} &= \text{according to equ. (6.5.1)} \\ \bar{a}, \bar{b} &= \text{constants.} \end{aligned}$$

6.5.1.5. To determine the variances of  $S$ , the type of distribution had to be taken into account. It can be shown by several tests that the distribution of the elements  $X_{ik}$  around their true, unknown mean value very closely follows a Poisson distribution. Two facts are remarkable in connection with the Poisson distribution:

i. The variance  $\sigma_i^2$  is given by:

$$\sigma_i^2 = \mu_i$$

ii. For  $\geq 30$  events (pulses) per time interval, the Poisson distribution can be approximated sufficiently well by a normal distribution, i.e.:

$$X_{ik} \sim n(\mu_i, \sigma_i) \quad (6.5.3)$$

Hence, as  $\geq 250$  events per time interval were measured in this case the estimates of  $\mu_i$  are given by the  $\bar{X}_i$  and the variance of these estimates is given by:

$$S_{\bar{X}_i}^2 = \bar{X}_i / 5 \quad (6.5.4)$$

Due to the property of normal distribution of Equ. (6.5.3) the

$S_{\bar{X}_i}^2$  are distributed:

$$\frac{5 S_{\bar{X}_i}^2}{\sigma_{\bar{X}_i}^2} = \chi^2 \quad (4) \quad (6.5.5)$$

With Equ. (6.5.5) an upper boundary at the 95 % confidence level can be calculated for the unknown  $\sigma_{\bar{X}_i}^2$  with respect to (6.5.4) :

$$\sigma_{\bar{X}_i}^2 = \frac{\bar{X}_i}{2} \chi_{0.05,4}^2 \quad (6.5.6)$$

With Equ. (6.5.6), one part of the variance of S has been established. The other contribution stems from the fact that  $\bar{a}$  and  $\bar{b}$  in Equ. (6.5.2) are realizations of random variables, too. Hence:

$$\sigma_S^2 = \sigma^2(\bar{a} \bar{X}) + \sigma^2(b) + \text{cov}(\bar{a} \bar{X}, \bar{b}) \quad (6.5.7)$$

In general, the covariance term in (6.5.7) has to be taken into account, but it can be shown that this term is not significantly positive in this case, hence:

$$\sigma_S^2 \leq \sigma^2(a \bar{X}) + \sigma^2(b) \quad (6.5.8)$$

The following equation for  $\sigma_S^2$  was used:

$$\sigma_S^2 = \sigma \frac{2}{b} + (\bar{a} \bar{X})^2 \left( \frac{1}{\chi^2 \cdot \bar{X}} + \frac{\sigma \frac{2}{a}}{-2} \right) \quad (6.5.9)$$

The values for  $\bar{a}$ ,  $\bar{b}$ ,  $\sigma \frac{2}{a}$  and  $\sigma \frac{2}{b}$  were calculated with the help of the "least squares" method.

With these values,  $\sigma_S^2$  could be calculated as a function of the  $\bar{X}$  observed. Typical values for

$$\delta_S = \sigma_S / S \quad (6.5.10)$$

are presented in Table XVII.

The function  $\delta(S)$  decreases with increasing S, a result which could be expected.

## 6.5.2 Measurements

For all batches  $i$  of the waste stream the pulses and the background were counted 5 times. With these measurements the respective quantities  $\bar{X}_i$  could be calculated. Then, the Pu-content was determined with Equ. (6.5.2). The respective Pu-content and the variance of the batches  $i$  can be calculated by:

$$m_{Pu_i} = \bar{C} \cdot S_i \quad (6.5.11)$$

where

$\bar{C}$  = the concentration determined in 6.5.1.3.

The coefficient of variation of  $m_{Pu}$  is given by:

$$\delta_{m_{Pu}}^2 = \delta_c^2 + \delta_S^2 + \delta_c^2 \cdot \delta_S^2$$

$$\sigma_{m_{Pu}}^2 = \delta_{m_{Pu}}^2 \cdot m_{Pu}^2 \quad (6.5.12)$$

where

$\delta_c^2$  = the coefficient of variation according to 6.5.1.3

$\delta_S^2$  = the coefficient of variation according to 6.5.10.

The values for  $\delta_{m_{Pu}}^2$  for different amounts of Pu are presented in Table XVI. From Table XVI one can see, that the influence of  $\delta_{\bar{C}}^2$  can be neglected.

The total amount of Pu contained in the whole waste stream was then calculated by

$$M_{W \text{ tot}} = \sum_{i=1}^K m_{Pu_i} \quad K = \text{all the batches containing waste}$$

and the variance

$$\sigma_{W \text{ tot}}^2 = \sum_{i=1}^K \sigma_{m \text{ Pui}}^2 \quad (6.5.13)$$

where

$M_{W \text{ tot}} \text{ [g]}$  = the total amount of Pu in the waste stream

$\sigma_{W \text{ tot}}^2 \text{ [g}^2\text{]}$  = the variance of  $M_{W \text{ tot}}$ .

These values were determined as:

$M_{W \text{ tot}} = 127.95 \text{ gms of Pu}$

$\sigma_{W \text{ tot}} = 1.49 \text{ gms of Pu.}$

The calculation of the material balance and the evaluation of the statements are described in 3.2.

TABLE I. SAFEGUARD MEASURES FOR THE CARRYING OUT OF THE MATERIAL BALANCE

Stream	No.	Measure
Input	M/1, M/2 M/3, M/4	Weighing, sampling and chemical analyses of the samples
Output pellets	M/5, M/6	$\gamma$ -spectroscopy and chemical analyses
Output pins	M/7	Calorimetry
Waste	M/8	Neutron counting
Scrap	M/1, M/2 M/3, M/4	Homogeneizing, weighing, sampling and chemical analyses

TABLE II. CONTAINMENT MEASURES

Location	No.	Typ	Measure
Pu-Storage	C/1	Sealing	Sealing of the Pu-Storage
Passage from Box CK 06 to CK 07	C/2	Sealing	Sealing to avoid transfer of not measured pellets
Calorimetry	C/3	Sealing	Marking of the pins measured to avoid recycling
Waste after measurement	C/4 + C/5	Sealing	Sealing to avoid recycling
Input and output streams	C/6	Identification	Mass-spectrometrical analyses

TABLE III. SPECIAL MEASURES

Location	No.	Measure
Between Pu-scrap and Box 1/85	S/1	Observation of the transports, to guarantee that the streams attained Box 1/85
Between Pu-storage and room 60 (calorimetry) and vice versa	S/2 S/3	Observation of the transports of the pins to avoid exchange of the pins
From room 89 to U-storage	S/4	Observation of the transports of recoverable waste

TABLE IV. LIST OF THE STRATEGIC POINTS

No.	Location	Function
1a	Pu-storage	Input and scrap
1b	Box 1/85	analysis. Safing of the product
2	Box CK 07/C1	Analysis of the product (pellet) stream by $\gamma$ -spectroscopy
3	Room 60	Analysis of Pu product (pin) stream by calorimetry
4	Room 89	Analysis of the waste stream by neutron counter

TABLE V: SAFEGUARD MEASURES AT THE FIRST STRATEGIC POINT  
(Pu-storage and box 1/85)

Place	Measure	No.	Principle function	Practical function
Pu-Storage	Sealing	C/1	Containment	Safing of input and scrap material
From Pu-storage to box 1/85	Transportation (Observation)	S/1	Special	To guarantee that the input stream attained box 1/85
Box 1/85	Weighing	M/1	Material Balance	Measurements of input and scrap stream
Box 1/85	Sampling	M/2	Material Balance	Sampling of Pu-containing powder
IRCH <sup>(1)</sup>	Chemical Analysis	M/3	Material Balance	Chemical Analysis of the samples taken
IRCH <sup>(1)</sup> and TU <sup>(2)</sup>	Mass-spectr. Analysis	M/4 C/5	Material Balance Containment	Mass-spectr. analysis for the material balance and the identifying of the material

(1) Institute for Radio Chemistry

(2) Transurane Institute

TABLE VI: SAFEGUARD MEASURES AT THE SECOND STRATEGIC POINT  
(CK 07/C1)

Place	Measure	No.	Principle function	Practical function
CK 07/C1	$\gamma$ -spectroscopy	M/5	Material balance	Measurement of the output stream by $\gamma$ -spectroscopy
CK 07/C1	Sealing	C/2	Containment	To avoid recycling of measured pellets
IRCh	Chemical Analysis	M/6	Material balance	Chemical analysis of the standard pellets

TABLE VII: SAFEGUARD MEASURES AT THE THIRD STRATEGIC POINT  
(Room 60: Calorimetry)

Place	Measure	No.	Principal function	Practical function
From Pu-storage to room 60	observation	S/2	special measure	to verify, that the pins measured could not be exchanged during transportation
room 60	calorimetry	M/7	material balance	measurement of the output stream by calorimetry
room 60	sealing	C/3	containment	to avoid recycling of measured pins
from room 60 to Pu-storage	observation	S/3	special measure	to verify additional to C/3 that no exchange or other manipulations were made with the pins during this transport

TABLE VIII: SAFEGUARD MEASURES AT THE FOURTH STRATEGIC POINT  
 (Room 89: Analysis of the waste stream)

Place	Measure	No.	principle function	Practical function
Room 89	n-counting	M/8	Material balance	Measurement of the Pu-content of the waste stream
Room 89	sealing	C/4	Containment	To avoid recycling of measured batches of waste
From room 89 to U-storage	observation	S/4	Special measure	To verify that the measured amounts of recoverable waste were transported to the U-storage
U-storage	sealing	C/5	Containment	Sealing of the past of the waste stream to be recovered to avoid recycling

TABLE IX : DATA OF THE PRODUCTION CAMPAIGN SAFEGUARDED  
DURING THE EXERCISE

1. General Data

Customer	United Nuclear Corp., N.Y.
Purpose	Fuel pins for the subcritical assembly PLATR
Amount of material delivered (shipper data)	ca. 5 kg of plutonium as PuO <sub>2</sub> ca. 250 kg of natural uranium as UO <sub>2</sub>

2. Specifications

Amount to be handled [ kg ceramic ]	200 kg [ U+Pu mixed oxide ]
----------------------------------------	-----------------------------

Concentration of Pu [ % ]	2.3
---------------------------	-----

Specifications of the pellets

Height [ mm ]	15
Diameter [ mm ]	12.5
Weight [ g, ceramic ]	18.6

Specifications of the pins

	Type I	Type II
Length [ inch ]	52.25	16.105
Number	113	73
Weight [ g ]	1472	452
Can. Material	Zircaloy II	

TABLE X. RESULTS IN % OF THE CHEMICAL ANALYSES OF THE SAMPLES WITH PU-CONCENTRATION OF CA. 87 % (INPUT)

Measurement No.	1	2	3	4
1	87.43	87.65	87.74	63.95
2	87.66	87.33	87.68	
3	87.24	87.23	87.60	63.96
4	87.37	87.56	87.36	63.71
5	87.54	87.20		
6	87.22	87.15		
$\bar{X}$ $\underline{[\%]}$	87.41	87.353	87.595	63.873
$s^2$ $\underline{[(\%)^2]}$	0.0244	0.0352	0.0208	0.01335
d $\underline{[\%]}$	0.1787	0.1877	0.1649	0.1809

TABLE XI. TEST FOR EQUAL VARIANCE (F-TEST) WITH THE RESULTS OF THE  
INPUT ANALYSES

Critical values for the ratio of the sample variances of the different  
batches at the 95 % confidence limit.

		Batch No.			
		1	2	3	4
	1	-	4.28	4.53	4.76
Batch	2	4.28	-	4.53	4.76
No.	3	6.16	6.16	-	6.59
	4	8.94	8.94	9.12	-

Actual values for the ratio of the sample variances.

		Batch No.			
		1	2	3	4
	1	-	1.442A	0.85A	0.54A
	2	0.693A	-	0.59A	0.379A
	3	1.173A	1.69A	-	0.642A
	4	1.82A	2.64A	1.55A	-

A: accepted as equal variance.

TABLE XII. TEST FOR UNCORRELATION OF THE CHEMICAL ANALYSES

Critical values of  $T_{ij}$  at the 95 % confidence level (two sided).

		Batch No. i			
		1	2	3	4
Batch No.	1		<u>+ 2.776</u>	<u>+ 4.303</u>	<u>+ 12.71</u>
	2			<u>+ 4.303</u>	<u>+ 12.71</u>
	3				<u>+ 12.71</u>

Actual values of  $T_{ij}$ 

		Batch No. i			
		1	2	3	4
Batch No.	1		+ 0.364 A	+ 1.251 A	+ 1.658 A
	2			+ 0.605 A	- 3.69 A
	3				+ 1.876 A

A: Uncorrelation accepted

TABLE XIII. RESULTS OF THE ANALYSIS OF VARIANCE

Batch No.	1	2	3	4
$\bar{X}_i$	87.41	87.353	87.595	63.873

Estimate for the variance:  $S_I^2 = 0.0320$

Degrees of freedom:  $f_I = 15$

TABLE XIV. RESULTS OF THE TEST OF THE HYPOTHESIS ABOUT THE VARIANCE OF THE CHEMICAL ANALYSES.

Critical value  $q_{crit}$  at the 95 % level:  
for 15.d.o. freedom

Upper bound:  $q_{crit o} = 25.0$

Lower bound:  $q_{crit n} = 6.26$

Actual values of  $q_i$ :

Batch No. i	1	2	3	4
$q_i$	2.62 R	2.63 R	3.92 R	9.81 A

A: Hypothesis accepted for coefficient of variation  $\delta = 0.2 \%$

R: Hypotheses rejected for coefficient of variation  $\delta = 0.2 \%$

Resulting coefficients of variations  $\delta_i$   
(see text)

Batch No. i	1	2	3	4
$\delta_i \left[ \frac{\%}{\%} \right]$	0.151	0.151	0.185	0.2

TABLE XV. ERROR IN THE MEASUREMENT OF CALORIMETRY ON ACCOUNT OF ISOTOPE MEASUREMENT ERRORS

Isotop.	perc.	error $\delta$ [%]	heat val. rel. w/g	watts
Pu <sub>38</sub>	0.27099	1.3	0.569	0.001542
Pu <sub>39</sub>	75.492	0.21	0.001923	0.0014517
Pu <sub>40</sub>	17.9703	0.56	0.00703	0.0012633
Pu <sub>41</sub>	4.8261	0.97	0.0045	0.0002172
Pu <sub>42</sub>	1.0704	1.33	0.00012	1.28 $10^{-6}$
Am <sub>41</sub>	0.3699	1.5	0.1084	0.000401
<b>Total:</b>		0.45		0.00487644 watt/gm of Pu safeguarded

TABLE XVI. TYPICAL VALUES FOR THE ERRORS OF THE CALORIMETRY  
DETERMINED DURING THE EXERCISE

a) Relative error of the line of equilibrium

$$\delta_{V_{R2}} = 1.1 \%$$

b) Relative error of the reference batch

$$\delta_{W_{AR}} = 0.1 \%$$

c) Relative error of voltage differences  $V_i$

$$\delta_{V_i} = 6.2 \%$$

d) Maximum relative error of the batches  $i$

$$\delta_{W_{A_i} \max} = 0.8 \%$$

e) Minimum and maximum error of the determination of Pu

$$\delta_{m_{Pui} \min} = 0.48 \%$$

$$\delta_{m_{Pui} \max} = 1.02 \%$$

TABLE XVII. COEFFICIENTS OF VARIATION FOR THE MEASUREMENTS OF THE WASTE STREAM AS FUNCTION OF THE PU-AMOUNT P

Pu-amount $\bar{g}$	0.2	0.5	1.0	10.0
Scrap $\bar{g}$ $\delta_S$	9.814	24.534	49.067	490.677
Coefficient of variation $\bar{\%}$ $\delta_S$	14.60	7.50	4.90	1.98
Coefficient of variation $\bar{\%}$ $\delta_{Pu}$	14.6	7.51	4.92	2.04

TABLE XVIII. RESULT OF THE MATERIAL BALANCE

Point	Amount $\bar{g}$ Pu $\bar{g}$	Standard deviation
Input	+ 5070.25	5.86
Output Pellets -spectroscopy	- 4209.57	4.91
Output Pins calorimetry	- 4213.7	14.48
Output scrap	- 677.63	2.19
Output Waste n-count	- 127.95	1.49
Output final scrap	- 6.5	0.06
Result E = 48.38		8.09
(difference between input and output)		

TABLE XIX. RESULT OF THE MASS-SPECTROSCOPIC ANALYSIS OF THE FINAL SCRAP

	Isotopes in %			
	Pu-239	Pu-240	Pu-241	Pu-242
Final Scrap	83.59	13.10	2.76	0.56
PLATR	75.49	17.97	4.83	1.07
SNEAK	91.0	6.32	0.6	0.03

TABLE XX. STATEMENTS ABOUT DIVERSION ON THE BASIS OF THE MATERIAL BALANCE

Probability (likelihood) of diversion:  $P_D$

	$P_D$ [%]		
	95	99	99.9
Q = Amount [ $\bar{g}$ Pu $\bar{g}$ ] (lower limit)	35.1	29.6	23.4
Percentage of actual amount diverted	83.2	67.8	35.5
Actual amount diverted:			42 g Pu

Risk (probability) of detection:  $R_D$   
as a function of  $P_D$  and Q

Q [ $\bar{g}$ Pu $\bar{g}$ ]	$P_D$		
	90	95	99.0
10	99.6	98.95	94.8
15	98.0	95.44	54.4
20	92.36	85.7	65.2
30	58.31	43.64	20.1
35	33.7	21.8	7

TABLE XXI. EFFORT OF THE SAFEGUARD EXERCISE

## a. Effort in man hours for the different safeguard activities.

Location	Man hours	% of total	% of total without γ-spectroscopy
Pu-storage	32	3.5	7.6
Box 1/85	62	6.9	14.7
Waste analysis	121	13.4	28.8
γ-spectroscopy	484	53.5	-
Calorimetry	70	7.7	16.6
Waiting time	80	8.8	19.0
Miscellaneous	56	6.2	13.3
Total	905	100.0	100.0

## b. Effort on chemical analyses.

Purpose	No.	% of total	% of total without γ-spectroscopy
Input	8	20.5	61.5
γ-spectroscopy	26	66.7	-
Scrap waste	5	12.8	38.5
Calorimetry	0	0	
Total	39	100	100

TABLE XXI. EFFORT OF THE SAFEGUARD EXERCISE (ctd.)

## c. Effort on mass-spectrometrical analyses.

Purpose	Location	No.	%of total
Homogeneity tests	Input	12	35.3
Identification	$\gamma$ -spectroscopy	12	35.3
Identification	Scrap + waste	5	14.7
Calorimetry	Calorimetry	5	14.5
	Total	34	100

TABLE XXII. RELATIVE IMPORTANCE OF THE STRATEGIC POINTS

Strat. Point	Measurement of	$\sigma_i$	man hours	chem. anal.	mass-spectr. analyses	$A_i$	$I_i$	$Z_i$ [%]	$Z_i$ [%] without $\gamma$ -spectroscopy
1a	Input and	6.256	94	13	17	7430	539.0	27.0	39.0
1b	Scrap								
2	pellets $\gamma$ -spectr.	4.91	484	26	12	15280	606.0	30.6	-
3	pins calorimetry	14.5	70	0	5	2670	744.0	37.4	54.0
4	waste	1.64	121	5	5	4710	100.0	5.0	7.0

Fig. 1

Alkem Plant Layout and the Locations of the Strategic Points for the Safeguard Experiment

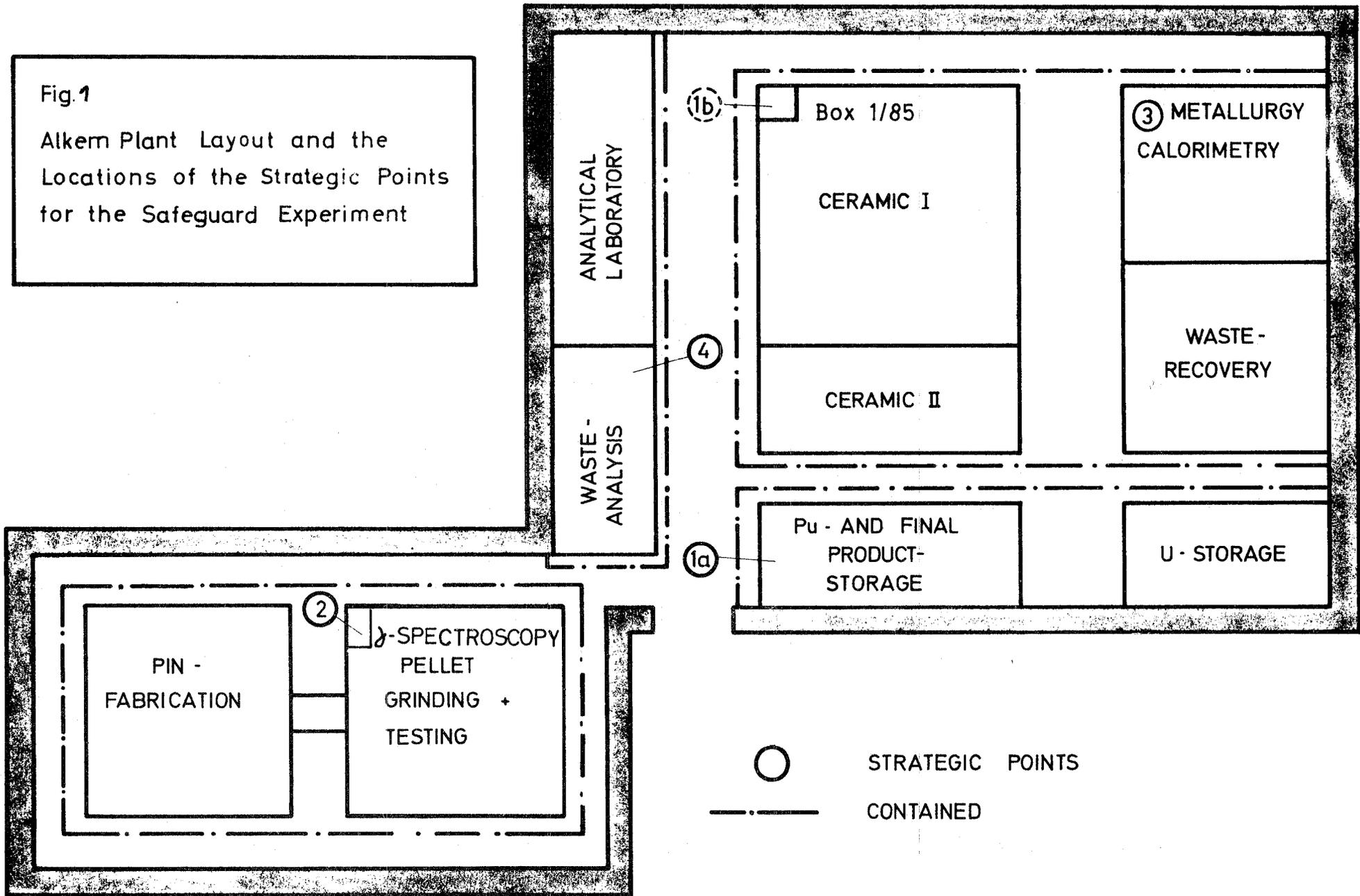


FIG.2 FLOW OF MATERIAL THROUGH THE STRATEGIC POINTS

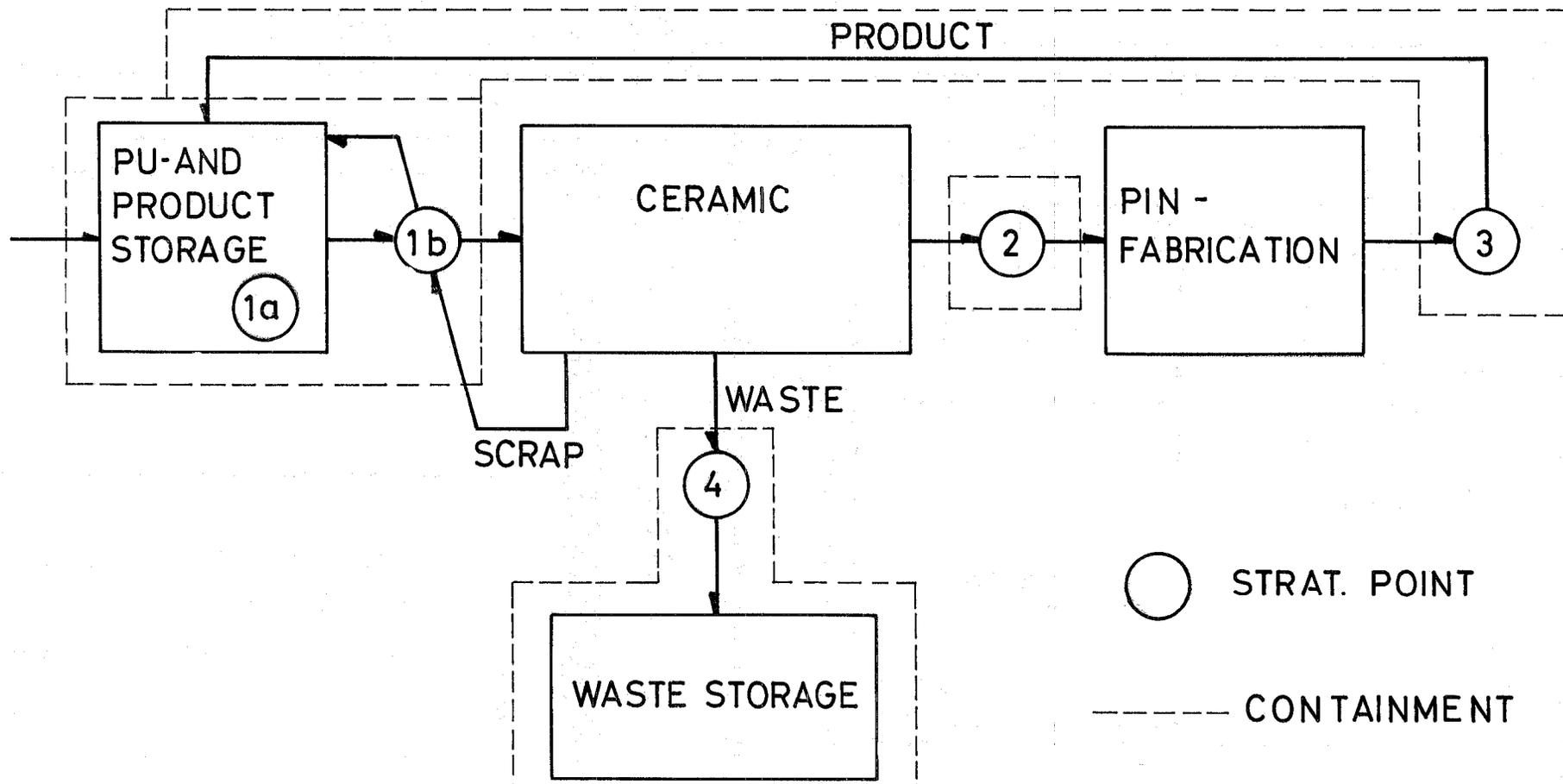


Fig.3 Arrangement of the  $\gamma$ -spektroskopy

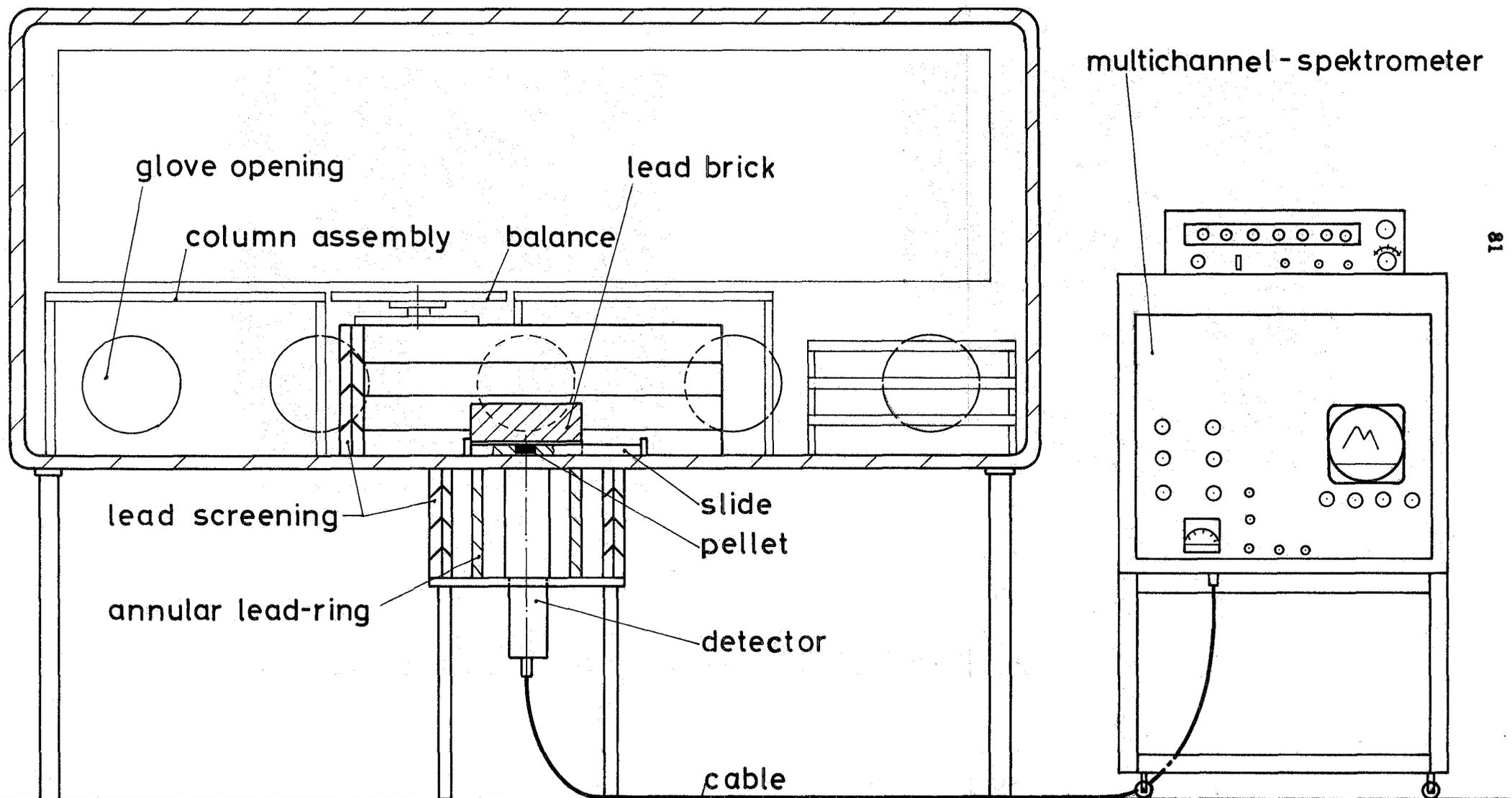
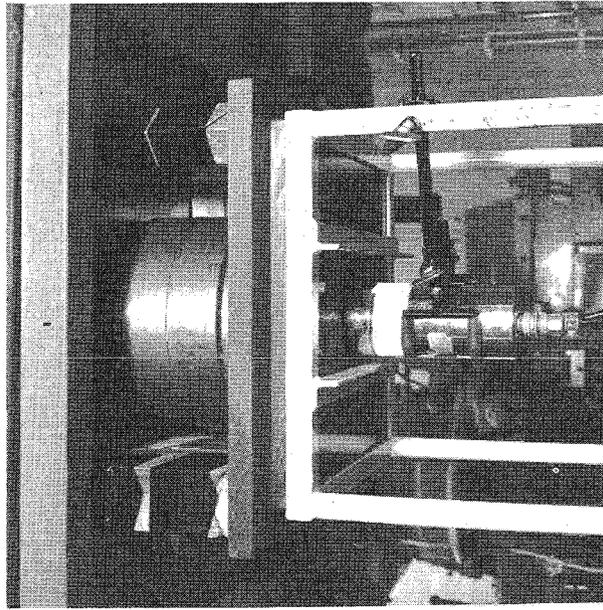
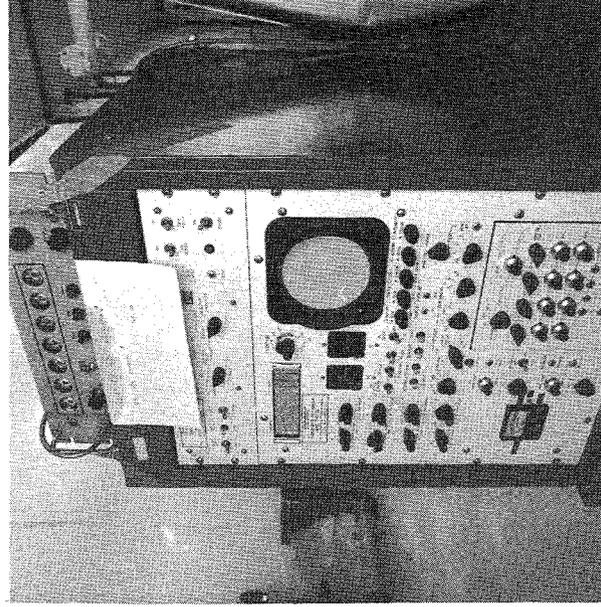


FIG 3 (CONTIN.) T-SPECTROMETER



DETECTOR



MULTICHANNEL WITH PEAK

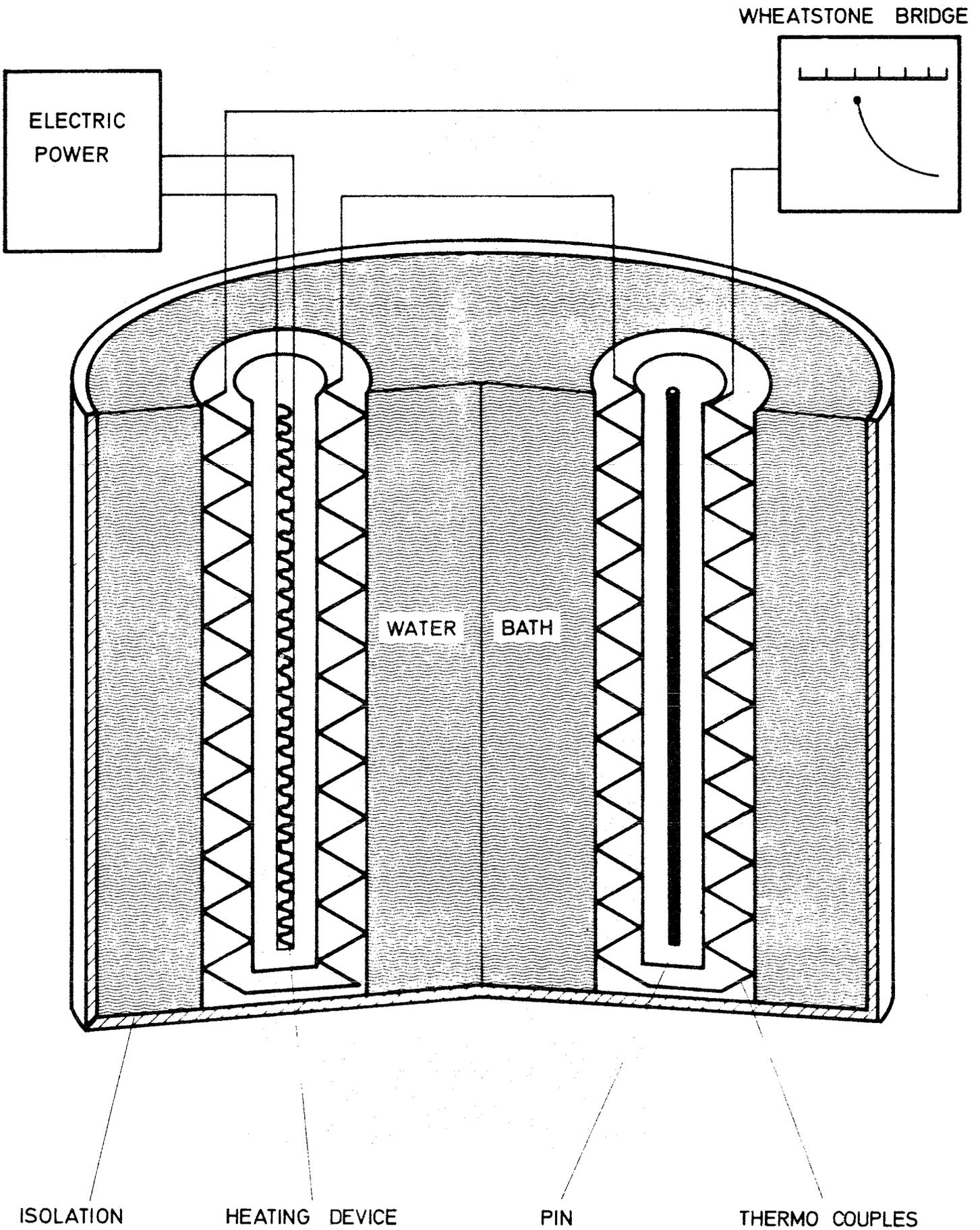
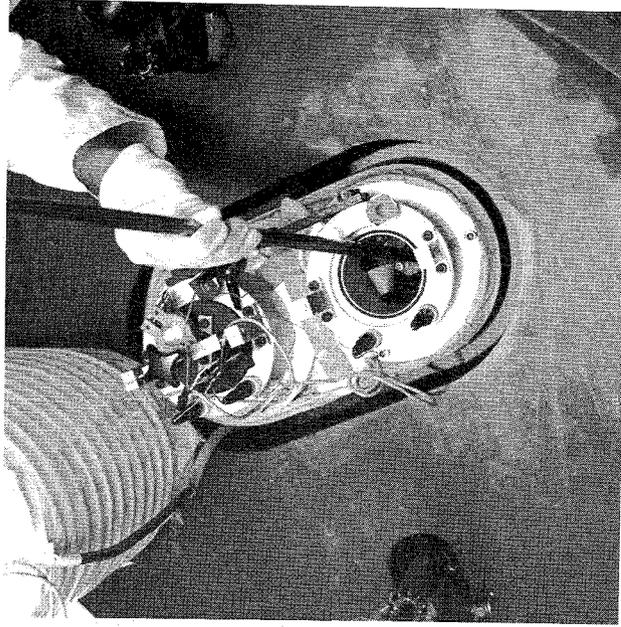
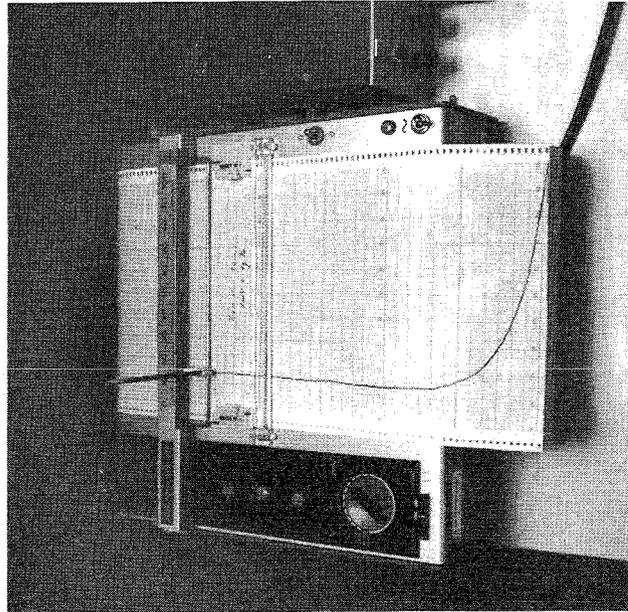


FIG. 4 PRINCIPLE OF THE CALORIMETER USED FOR THE SAFEGUARDS EXERCISE

FIG. 5 (CONTIN.) CALORIMETER

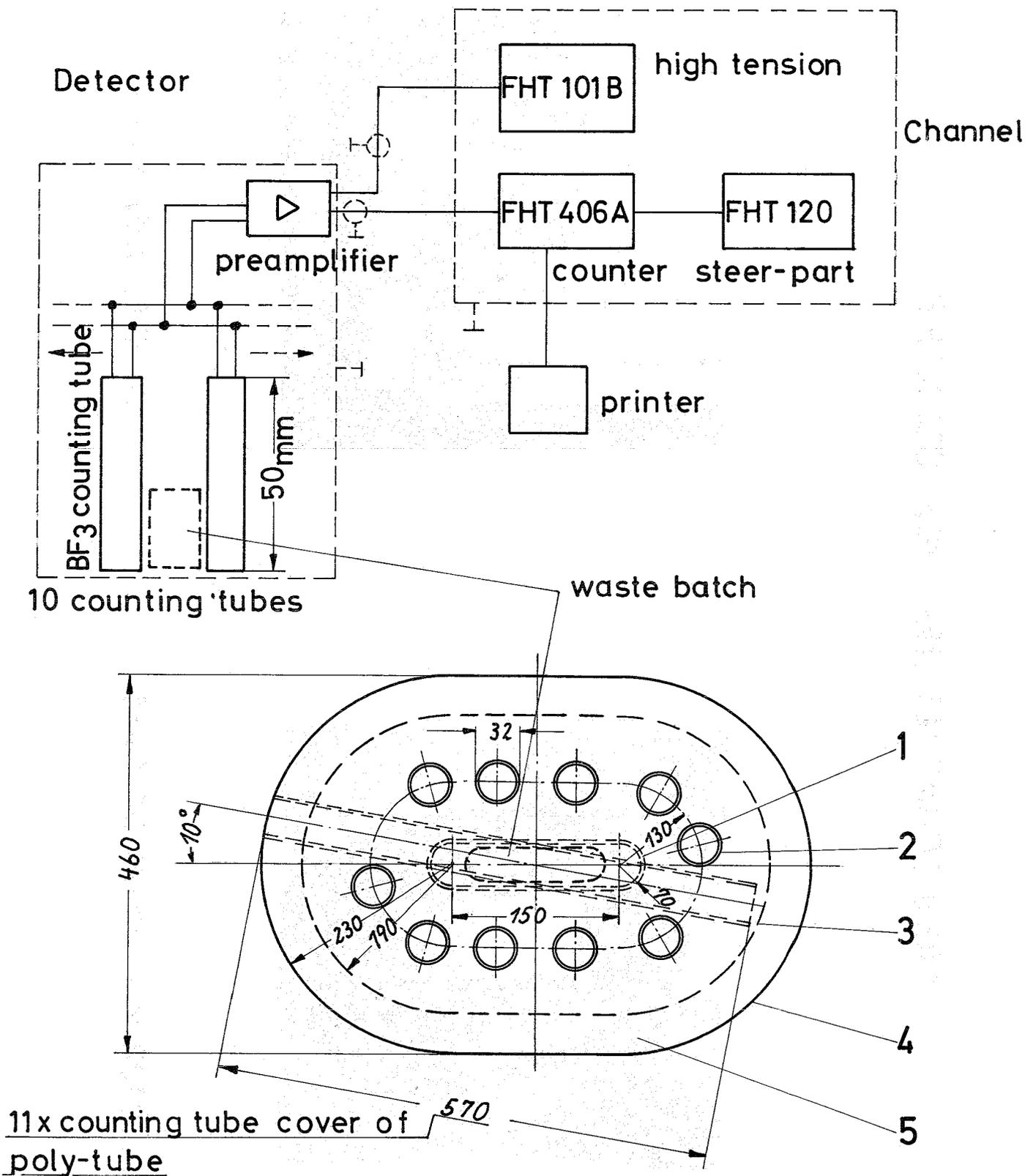


THE TWO CHAMBRES



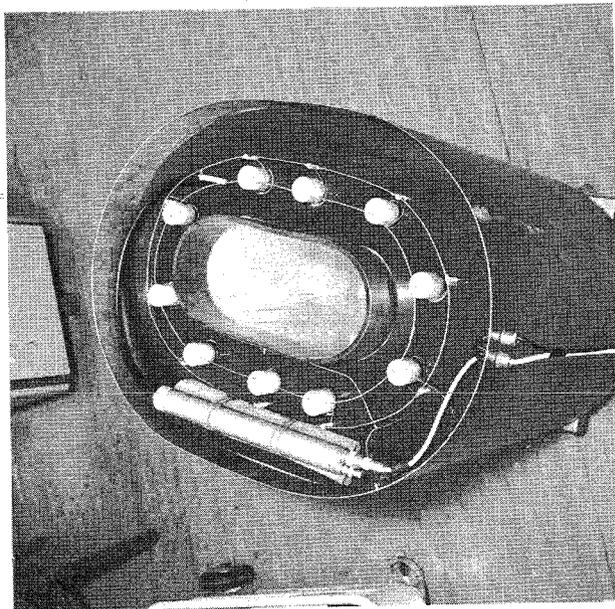
TYPICAL RECORDER CURVE

Fig.6 Principle of the neutron counter (multi channel)

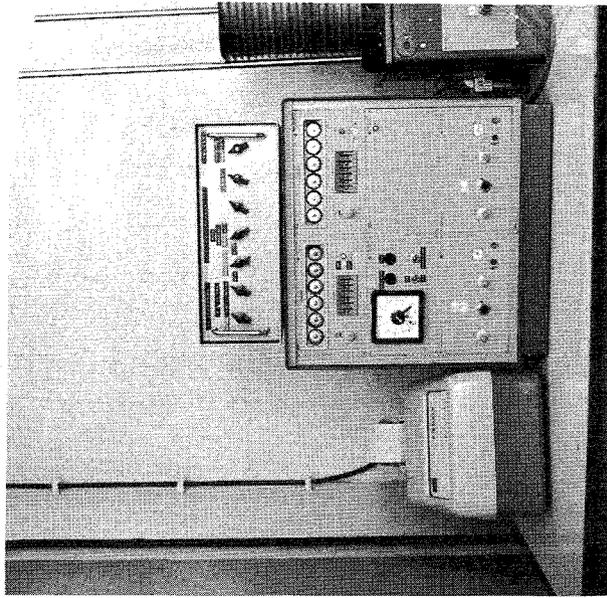


- 1 Plexiglass vessel wall thickness 5mm
- 2 Counting tube cover Polyethylene  $d_1=27\text{mm}$ ,  $d_2=32\text{mm}$
- 3 Sheet of cadmium 1mm
- 4 Cadmium-plated steel sheet 2-3 mm
- 5 Paraffin

FIG. 6 ( CONTIN.) NEUTRON COUNTER



DETECTOR WITH COUNTING TUBES



CHANNEL AND PRINTER

FIG.7 TIME SCHEDULE OF THE EXERCISE

PHASE NO.	MAY	JUNE	JULY	AUG.	SEPT.	OCT. 68
I	Arrival Training period					
II		Clean out				
III		Analysis of input stream n-Count standarts				
IV		Installation $\gamma$ -spectroscopy $\gamma$ -spectroscopy				
V	Final test period of calorimetry			Waste analysis	Calorimetry	Statistical analysis
					Scrap analysis	Final clean out
NUMBER OF PERSON.	4	4	3	2	2	1

# FIG. 8 EXAMPLES FOR CONTAINMENT (SEALING) MEASURES



PU - STORAGE



WASTE BARRELS

# FORMBLATT FC-ANS-2 (BOX 1/85 und BOX 2/61)

Phase Nr.	1) Datum	2) Uhrzeit	3) Auftrag	4) Materialtyp	Temperatur in Sox	Letzte Registrierung	5) Übergabe Mat. an BOX Nr./Raum Nr.	5) Übergabe Waste an BOX Nr./Raum Nr.	Brutto Wägung (g)	Leerbehälterwägung (g) tara	6) Nullpunkt kontr.	rel. Luftfeucht.	Unterdruck im Innern gegen Außenw.	7) Proben nr.	Analysen ergebnisse	Kommentar bzw. Dosen Nr. u.ä.	Blatt Nr.	Lfd. Nr.	Name
0.2	105.27	10.45	PPP	22	01	01	1 K08/85	K01/85	11.445	.	J	65	33	B			01	0	KR
0.2	205.28	11.20	LL	21	01	01	3 K01/85	K01/85	1.730	1.713	J	65	33	I 02		113	01	1	GM
0.2	05.28	11.20		22	01	01	4 PULAGER	K01/85	1.755	1.747	J	65	33	I 01		112	01	2	GM
0.2	05.28	11.30		22	01	01	4 K01/85	K01/85	12.529	10.036	J	65	33	B		112	01	3	GM
0.2	05.28	11.35		21	01	01	3 K01/85	K01/85	12.051	10.155	J	65	33	B		113	01	4	GM
0.2	05.28	11.45		22	01	01	4 K08/85	K01/85	2140.0	1040.2	J	65	33			112+PRDB.	01	5	GM
0.2	05.28	12.00		22	01	01	3 PULAGER	K01/85	1208.2	71.4	J	65	33			113+PRDB.	01	6	GM
0.2	05.28	14.30		22	01	01	7 PULAGER	K01/85	1199.0	66.3	J	65	33			110+PRDB.	01	7	GM
0.2	05.28	15.20		22	01	01	6 PULAGER	K01/85	1222.5	66.4	J	65	33			111+PRDB.	01	8	KR
0.2	05.28	14.30		22	01	01	7 K01/85	K01/85	12.309	10.324	J	65	33			110	01	9	KR

ERLÄUTERUNGEN

1) Datum

z.B. 4.5.68 zu schreiben: 0504

3) Auftrag Bezeichnung  
PLATR P  
Masurca M

2) Uhrzeit

931 zu schreiben: 0931

4) Materialtyp Bezeichnung  
Pu<sub>02</sub> Pulver PP  
Pu<sub>01</sub> U. Pulver UP  
Ausschuß Pellets AP  
Schleifschlamm SS  
Masurca scrap. MS  
Masurca Endprod. ME

5) z.B. CK 07/C1

6) Kontrolle vorgenommen

JA NEIN  
J N

7)

Nur auszufüllen bei Probenentnahme

8) Lfd. Nr. der letzten Eichung

Zweck Bezeichnung  
Proben Alkem BET B  
" IRCH " Analytik A<sub>I</sub>

FIG. 9. FORM FOR INPUT AND SCRAP ANALYSIS

BLATT NR. 7

MESSPROTOKOLL  $\gamma$ -MESSUNG  
FC-ANS-05

Blatt Gesamtzahl Pellets  
Nr.: Datum Uhrzeit von bis des Losses

Opera-  
teure

Kommentar

007 0718 1630 1730 157 STAND. BLATT 4 STANDART PELLLET NEDT

MESSERGEBNISSE											Kommentar	
Untergrund			Pellet			Untergrund			Pellet			
Nr.	Impulse	Nr.	Impulse	Impulse	Impulse	Nr.	Impulse	Nr.	Impulse	Impulse		Impulse
			1. Messung	2. Messung	3. Messung				1. Messung	2. Messung	3. Messung	
0 1	516	0 1	32261			0 2	496	0 2	33793			
0 3	525	0 3	32271			0 4	514	0 4	32337			
0 5	548	0 5	32336			0 6	504	0 6	32184			
0 7	520	0 7	32288			0 8	590	0 8	32401			
0 9	532	0 9	32419			1 0	574	1 0	32326			
1 1	597	1 1	32512			1 2	562	1 2	32266			
1 3	517	1 3	32172			1 4	545	1 4	32332			
1 5	531	1 5	32279			1 6	507	1 6	32425			
1 7	578	1 7	32545			1 8	554	1 8	32210			
1 9	528	1 9	32163			2 0	537	2 0	32430			
2 1	581	2 1	32399			2 2	538	2 2	32304			
2 3	558	2 3	32266			2 4	544	2 4	32170			
2 5	583	2 5	33090			2 6	558	2 6	33624			
2 7	575	2 7	32659			2 8	549	2 8	32170			
2 9	568	2 9	33267			3 0	584	3 0	33374			

Anmerkungen

Verlauf der Messung:

1. Das Los wird abgezählt
2. 30 Pellets werden abgesondert
3. Die 30 Pellets werden gemessen
4. Das letzte dieser 30 Pellets wird zurückbehalten
5. Das zurückbehaltene Pellet wird zusammen mit einem Zettel ausgeschleust und im Lager versiegelt.
6. Auf diesem Zettel steht: Blatt Nr., Datum, Gesamtzahl der Pellets