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NON-DESTRUCTIVE DETERMINATION OF FISSION GAS RELEASE BY GAMMMA SPECTROMETRY

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1. INTRODUCTION

In the frame of an experimental programme on LWR fuel transient behaviour, the determination of the fission product distribution as well as the fission gas release are key factors in understanding the overall behaviour of the fuel rod.

Two different methods for the determination of the fission gas release by gamma spectrometry of the ^{85}Kr isotope have been evaluated.
Further research is foreseen before a definitive choice for one of both methods can be made.

In the following, a description is given of the experimental configuration and data processing with some fuel rod test results.

2. EXPERIMENTAL CONFIGURATION

2.1. Apparatus - Metrology Bench

The gamma scanning operation is performed on the metrology bench (I) of the LHMA hot cell for non-destructive examinations.
For gamma scanning measurements, the main elements involved of this bench I are the following.

- A supporting table with a length of about 6.0 m backed up by an extension tube fitted with fuel rod supporting elements allowing scanning translation to be made of fuel rods having a maximum length of about 4.5 m.

- A trolley, driven by a stepping motor system, under computer control, allowing the accurate translation of the fuel rod under examination.
  On this trolley, a chuck for positioning, clamping of the fuel rod end cap and rotation of the rod by a stepping motor drive system (computer-controlled) is foreseen.

- Roller supports fitted on the table and in the extension tube for fuel rod loading over the entire length.

- Fuel rod guides fitted on the table and positioned in front and behind the slit of the collimator.

- A collimator : on the floor of the cell, a shielded cylindrical drum housing the collimating system has been mounted. Different exchangeable collimators precisely define the specified area of the irradiated fuel pin under examination.
  These primary collimators, composed of a denal insert (tungsten alloy) surrounded by lead for shielding, have different slit dimensions.
  The rotating drum allows the selected slit to be assigned. A small vertical translation of the collimator has also been foreseen. All movements are stepping motor driven under computer control.

The detector system (Ge), positioned in the basement underneath the hot cell is mounted on a yoke which may be moved such as to be capable of aligning the detector shielding and the outer slit collimator.

Fig. 1 illustrates the detection system.

2.2. Analytical Instrumentation

The analytical instrumentation is composed of the following units :

- Germanium detector (model GC 1018-7500) with following specifications :
  - * resolution 1.8 KeV (FHHM) at 1.33 MeV
  - * relative efficiency 10%
- Pre-amplifier Model 2001
- Fast spectroscopy Amplifier Model 2024
- S80 Multi Channnel Analyzer with 4095 channels
- PDP11 Computer to analyse the spectra collected with the S80 MCA
  - Consist of : * Video terminal VT220
    - * PDP 11/03
    - * Proprinter
    - * Hp 7475A Plotter
    - * Hard disk
    - * Floppy disk
- The Noron-computer to positioning the specimen on bench 1 of the hot-cell.

3. METHOD AND EVALUATION TECHNIQUE

3.1. General

85Kr is the only isotope of the noble gases that is produced during fission with a half-life that is high enough (10.76 year) and is therefore the only candidate to use for non-destructive examination of released fission gases by means of gamma spectrometry.
The main gamma peak of 85Kr is at 514 keV. This is a rather troublesome position, since it is very near to the annihilation peak at 511 keV and the 512 keV gamma peak of 106Rh. Especially the vicinity of the 512 keV 106Rh peak influences the precise determination of the peak area of the 514 keV peak. An example is shown in figure 3.
Another point that influences the precise determination of the 514 keV peak area is the presence of the cladding and the spring in the gas plenum. Activation of the spring and of cladding material will cause an elevated Compton level at the 514 keV peak. Figures 4a and 4b show measurements of 85Kr in a specially constructed gas chamber without spring and a common gas plenum with spring, clearly indicating that activation of the spring gives the highest contribution to the elevated Compton level in comparison with activation of the cladding.

3.2. Punction and mass spectrometry

In order to determine accurately the amount of fission gases that has been released from the fuel pellets one has to perform a punction in the gas plenum of the specific rod. The released fission gases are expanded into a well-known volume and with help of a simple determination of the gas pressure the volume (at STP) of released fission gases can be determined.
With help of mass spectrometry the relative fractions of the different fission gases (Kr and Xe) and their respective isotopes can be determined. In this way the amount of 85Kr in the gas plenum of a fuel rod can be determined.

Since the relative amount of 85Kr with respect to the other fission gases is rather low, the mass spectrometric results suffer from rather high uncertainties with respect to the 85Kr content.

3.3. Comparative non-destructive measurements

If a punction cannot be performed (e.g. if further irradiation of the rod is foreseen), one cannot use this method to determine the released amount of fission gas. More indirect methods have to be used. At the SCK/CEN two rather similar ways have been followed to determine non-destructively the fission gas release in a fuel rod. Both methods are based on a measurement of the emitted 514 keV gamma photons from 85Kr.

For the first way three experimental rods have been used. Apart from the usual gas plenum with spring at the upper side of the fuel pin, these experimental rods also have a gas chamber at the bottom side of the fuel pin without a spring. This gas chamber has been provided in order to reduce the Compton level caused by activation of the spring (figure 4a and 4b).
These experimental rods have been irradiated together. All rods have been examined non-destructively with respect to the presence of the 514 keV gamma peak of 85Kr and two out of three have been punctured for an absolute determination of the 85Kr content (and all other fission gas isotopes) by expansion into a well-known volume and subsequent mass spectrometry analysis. One rod is kept in its former condition in order to serve as a monitor rod for future measurements. Due to the determination of the 85Kr gamma peaks a direct relationship has been found between the 85Kr content of the monitor rod and the punctured rods.
Due to the reduction of the Compton level the amount of 85Kr in the monitor rod has been determined more accurately than one could have done in a normal rod without gas chamber. Therefore it can be stated that the monitor rod is better qualified to perform comparative measurements than a normal rod without a gas chamber.

It should be noted that comparative measurements on the monitor rod and a test rod of standard fabrication should both be executed on the gas plenum with the spring. In this way deviations due to the shielding correction for the spring and eventually due to a high Compton level will tend to be cancelled out, while otherwise the shielding correction for the spring will only be applied to the test rod, which could cause a deviation only on the results for the test rod.
Each time a punction has to be performed on a rod is another occasion to do a calibration of the monitor rod. The more often a calibration is performed, the better the overall calibration of the monitor rod will be. So in the course of time we will be able improve the results of our monitor rod with respect to the precision in the determination of the 85Kr content of fuel rods.

The second way is to irradiate a series of rods and to perform non-destructive 85Kr measurements on each of them. Several rods are then punctured and inspected by mass spectrometry in order to determine the absolute amount of 85Kr that has been released. By comparison one can determine the content of 85Kr in the gas plenum in the rods that have not been punctured. Taking the ratio between 85Kr and total Kr and the ratio between Kr and Xe as determined by mass spectrometry, one can deduce the amount of released fission gases in the fuel rod.
In fact this is the same as the above mentioned method, except that one does not use a specially constructed monitor rod and that one has to redo the calibration each time one measures a new series of rods.
A disadvantage of this method is that one cannot improve the precision of the calibration since one starts from zero each time a new series of rods is measured.

3.4. Gamma peak evaluation

An important aspect of the evaluation of the measurement results is the gamma peak evaluation. The area of the gamma peak is determined by fitting a Gaussian curve on a polynomial underground. Normally a first order polynome is taken, i.e. a

straight line, since this gives the most satisfactory results. Due to the vicinity of the 512 keV peak of 106Rh and to a lesser extent the annihilation peak at 511 keV, one is obliged to fit the regarded energy interval with multiple Gaussians in order to account for the presence of these peaks. An uncertainty analysis of the fitting procedure is part of the evaluation.

4. MEASUREMENTS IN PRACTICE

4.1. Non destructive measurements with monitor rod

In the past several measurements have been performed in order to calibrate a monitor rod with respect to its 85Kr content of the gas plenum. This monitor rod will be used in future comparative measurements to determine in a non destructive way the 85Kr content in an irradiated fuel pin.
The calibrations consisted of non destructive measurements of the 85Kr isotope by gamma spectrometry on several fuel pins, especially constructed with a gas chamber (see figure 5). All but one fuel pins have been punctured thereafter in order to determine the absolute amount of 85Kr released in the fuel pin with the help of mass spectrometry. In this way the 85Kr content of the non-punctured rod has been determined by comparing the results of the non destructive measurements and the punction. The results of these measurements are listed in table I. It appears that the measurements agree within an interval of approximately 5%. The non-punctured rod will be used further as a monitor for comparative 85Kr measurements. Should the occasion occur, more calibrations of this rod will be performed in order to obtain a still better value for its 85Kr content.

Table I.
Results of the calibration of the 85Kr content of the monitor rod F-1077. The measured values of rod F-1066 have been taken as reference. Rod 1-329 has been measured twice by gamma spectrometry. The relative difference has been taken with respect to the mean.

rod	ratio gamma spectrometry	ratio mass spectrometry	relative difference
monitor rod F-1077	1.5349		
F-1066	1	1	4.5%
1-329 (I)	1.8855	1.7053	- 5.3%
1-329 (II)	1.8327	1.7053	- 3.7%

Several contributions to the measurement uncertainty can be distinguished:

- uncertainty in the mass spectrometric determination of the Kr content of the released fission gas
- uncertainty in the mass spectrometric determination of the 85Kr content of Kr
- uncertainty in the determination of the free volume of the monitor rod
- uncertainty in the determination of the free volume of the reference rods
- uncertainty due to counting statistics
- uncertainty in the gas volume determination of the reference rod

The respective values of these contributions are listed in table Ib.

Table Ib.
Most important contributions to the uncertainty of the calibration of the monitor rod.

contribution	uncertainty (1 sigma)
mass spectrometry on Kr	5-10% *
mass spectrometry on 85Kr	10% *
free volume monitor rod	4.5%
free volume reference rod	3.5%
counting statistics	2%
gas volume reference rod	1%

* Dependent on the content

4.2. Non destructive measurements without monitor rod

Lately a series of 8 rods has been measured non destructively by gamma spectrometry and afterwards 6 of them have been punctured in order to determine the content of Kr and Xe isotopes. The results of these measurements are shown in table II. These measurements agree within 13%.

Table II.
Results of measurements with gamma spectrometry and mass spectrometry. Rod Pu6 did not contain a significant amount of fission gas in the gas plenum. The relative difference has been taken with respect to the mean.

rod	ratio gamma spectrometry	ratio mass spectrometry	relative difference
Pu1	1	1	1.6%
Pu2	3.7169		
Pu3	2.6182	2.8878	11.9%
Pu4	1.9814	2.0224	3.7%
Pu5	4.6443		
Pu6	0	0	
U1	1.1425	1.0000	-12.7%
U2	3.6685	3.4519	- 4.7%

The contributions to the measurement uncertainty can be distinguished as follows:

- uncertainty in the mass spectrometric determination of the Kr content of the released fission gas
- uncertainty in the mass spectrometric determination of the 85Kr content of Kr
- uncertainty in the determination of the free volume of the non-punctured rods
- uncertainty in the determination of the free volume of the punctured rods
- uncertainty due to counting statistics
- uncertainty in the gas volume determination of the punctured rods
- uncertainty due to shielding correction

The values of these contributions are listed in table IIb.

Table IIb.
Contributions to the uncertainty in the determination of the 85Kr content of the irradiated fuel rods.

contribution	uncertainty (1 sigma)
mass spectrometry on Kr	5-10% *
mass spectrometry on 85Kr	10% *
free volume of the non-punctured rods	3%
free volume of the punctured rods	3%
counting statistics	3%
gas volume of the punctured rods	3%
shielding correction	3%

* dependent on the content

5. QUALIFICATION

5.1. Punction and mass spectrometry

A punction together with mass spectrometry offers the most precise way to determine the amount of released fission gases in a fuel rod. The uncertainty is approximately 3.3% and is mostly due to the uncertainty in the determination of the gas volume.
Due to the rather large uncertainties in the relative presence of 85Kr by mass spectrometry, one could ask oneself if it should not be worthwhile to replace also these measurements by gamma spectrometry. However, one should keep in mind that the final objective of these measurements is not the 85Kr content of a fuel rod, but the total content of released fission gases. The total content can be measured very precisely by a punction combined with mass spectrometry (or a chemical assay of the content of the gas chamber).

5.2. Non destructive gamma spectrometry

The results of the calibration measurements thus far indicate that the spread of the measurements with the monitor rod is smaller than the spread of the measurements with the series of 8 rods. Since very few measurements have been performed with the monitor rod, one has to be very careful for making any conclusions at this point. However, it is clear that the uncertainties for both methods are of the same order of magnitude.
Since each recalibration of the monitor rod will reduce the uncertainty with respect to the 85Kr content of the monitor rod, it can be expected that in the course of time the monitor rod will offer a better way to determine the amount of 85Kr in an irradiated fuel rod in a non destructive way.

In order to obtain the best possible results with the monitor rod one should take into account the following:

- comparative measurements on the monitor and a test rod

have to be performed at the gas plenum with the spring. An additional measurement on the special gas chamber of the monitor rod can be carried out, but the result should not be used in the comparison, since the shielding corrections in this case will be rather different.

- Since a well-known calibration source is available, it is sensible to perform all measurements together with a measurement of this Eu calibration source, in order to detect in an early stage deviations in e.g. the detector-source geometry.

- In order to allow an as good as possible distinction between the different gamma peaks, the measurements have to be performed with many channels in the neighbourhood of the energy of 514 keV.

A point of uncertainty for both non destructive methods is the fact that one has to make an assumption with regards to the ratio of Xe and Kr and the ratio of 85Kr and total Kr.
The Xe/Kr ratio depends heavily on the type of fuel. For the 85Kr/total Kr ratio the situation is more complicated. Table III shows some interesting data with respect to this point.
From the data shown in the table it has been assumed that the 85Kr/total Kr ratio is not dependent on the type of fuel, but depends rather strongly on the irradiation conditions. All rods that have been irradiated in reactor 1 have been irradiated during the same cycles and show no large deviations in the 85Kr/total Kr ratio, independent of the type of fuel. The same accounts for the fuel rods irradiated in reactor 2. However, dependent on the reactor in which the rods have been irradiated, a large difference in the ratio can be observed (approximately 28%).

Table III.
Xe/Kr and 85Kr/total Kr ratios for several punctured fuel rods.

rod	irradiated in	Xe/Kr	85Kr/total Kr
Pu1	reactor 1	15.1	0.0732
Pu3	reactor 1	14.5	0.0732
Pu4	reactor 1	14.7	0.0745
F-1066 (Pu)	reactor 2	15.5	0.0583
1-329 (U)	reactor 2	7.26	0.0582
U1	reactor 1	6.95	0.0741
U2	reactor 1	7.56	0.0740

Since the number of measurements is very small, one has to be be careful in drawing too definitive conclusions. But on basis of the data listed in table III one could preliminarily state that the Xe/Kr ratio depends in first order on the type of fuel (Pu or U). The 85Kr/total Kr ratio does not depend on the type of fuel, but in first order on the irradiation conditions. No conclusions can be drawn with respect to what parameter is important for the 85Kr/total Kr ratio (neutron spectrum, maximum temperature, burn up, irradiation profile?).

The foregoing shows clearly that additional calibration measurements are necessary in order to determine the cause of the different 85Kr/total Kr ratio and to verify the constant Xe/Kr ratio (for Pu and U, resp.). Until the 85Kr/total Kr matter has been resolved, each non destructive measurement on fuel, irradiated under new circumstances, should be accompanied by a mass spectrometry measurement in order to verify the 85Kr/total Kr ratio.

5.3. Peak evaluation

Due to the use of a less qualitative Ge-crystal one has been obliged to adapt the gamma peak evaluation methods in order to account for lower tailing of the gamma peaks. This adaptation had not been performed at the time that the described measurements were evaluated, but due to the low counting rate and the consequent rather low statistics the lower tailing was not of great importance for these measurements.

6. CONCLUSIONS

Mass spectrometry together with expansion into a well-known volume is the most precise method in order to determine the total amount of released fission gases in a fuel rod. Since this is a destructive method, it cannot be applied in case the investigated fuel rod has to be used in future experiments.

The monitor rod offers the best possibility for future non destructive determination of the fission gas release in an irradiated fuel rod. At performing measurements one has to take into account the following:

- perform measurements on gas plenum with spring
- eventually incorporate measurement on Eu calibration source
- measure with many channels in the energy range of 514 keV

The dependence of the 85Kr/total Kr ratio on the irradiation conditions makes it necessary that further calibrations of the monitor rod are performed in order to verify for each type of irradiation the 85Kr/total Kr ratio.
From these calibrations it can probably appear that one mass spectrometric measurement is to be incorporated with the non destructive method when measuring fuel rods with the same irradiation history.

Due to the small relative content of 85Kr in the fission gases the determination of the 85Kr content by mass spectrometry is rather imprecise. The 85Kr determination will improve for higher fission gas releases.

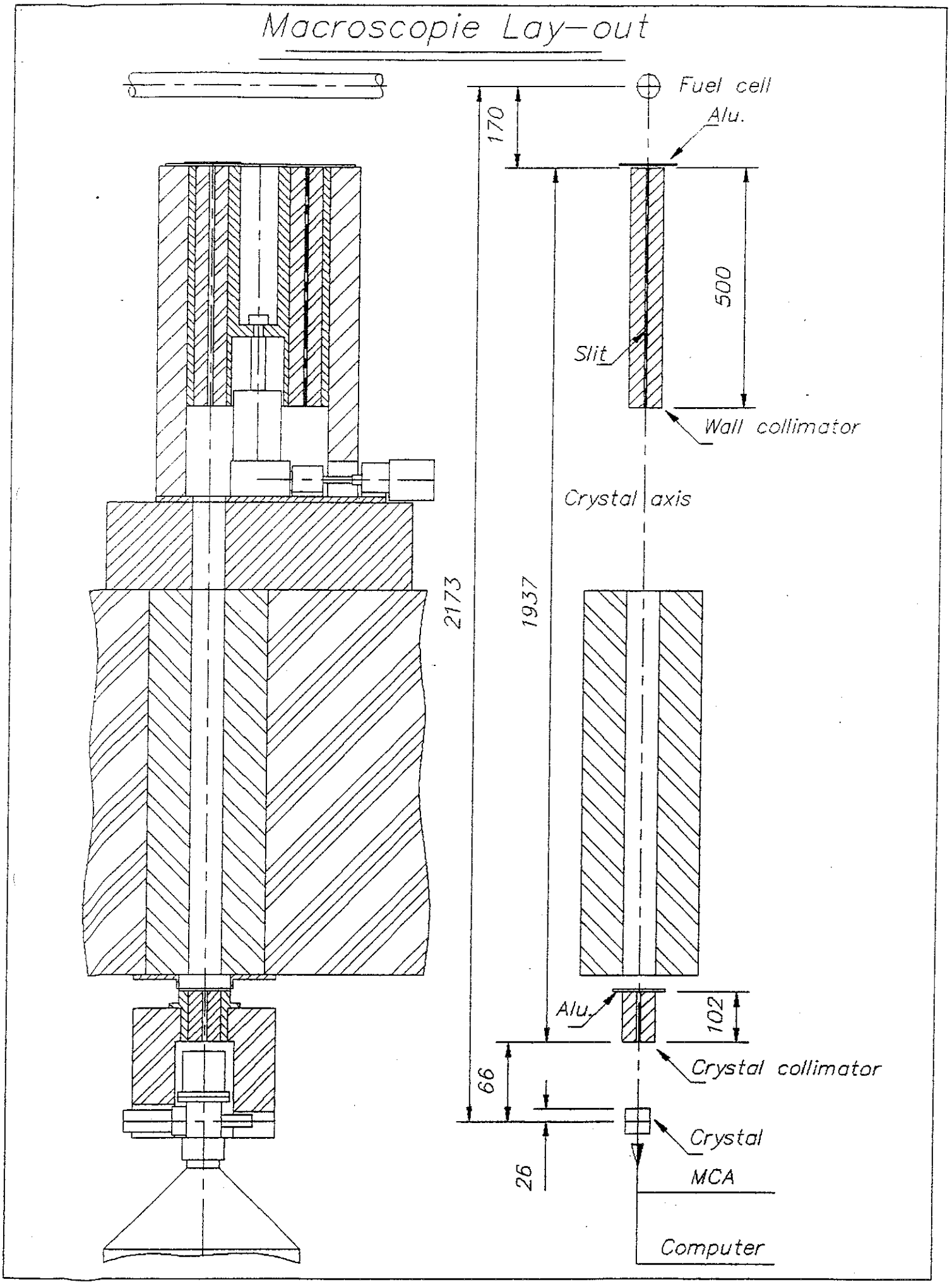
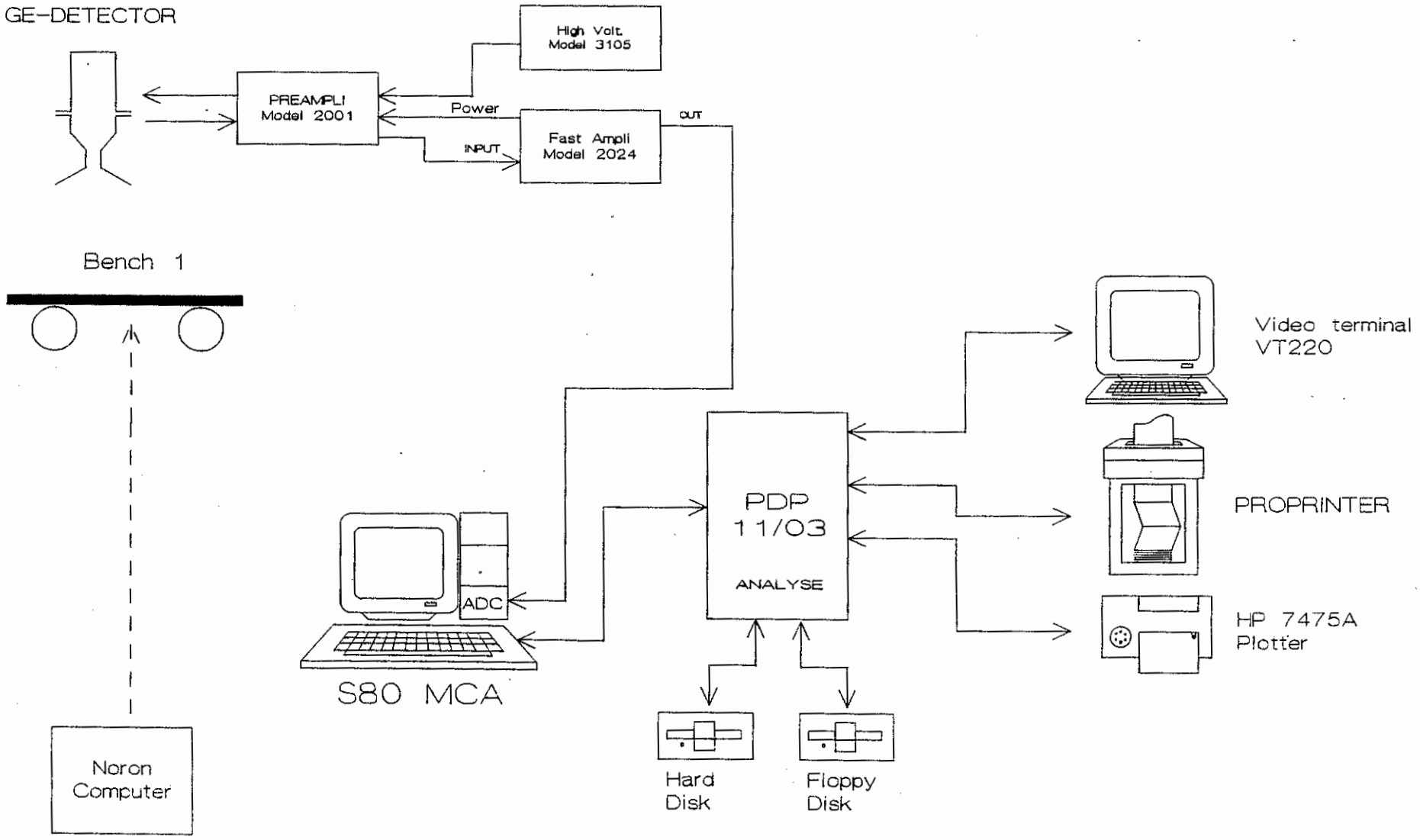


Fig. 1. Macroscopic lay-out of the activity measurement system.

Fig.2. HIGH RESOLUTION GAMMA-SPECTROMETRY



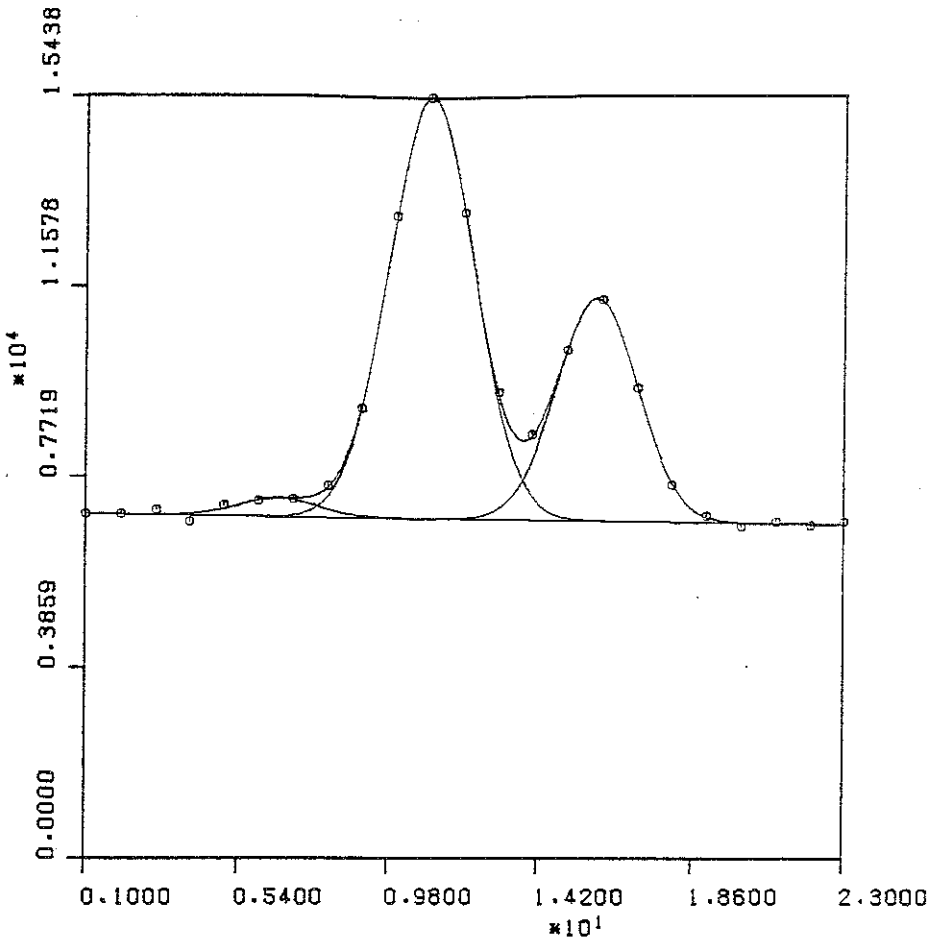


Fig. 3. Example of annihilation peak (511 keV), ^{100}Rh peak (512 keV) and ^{85}Kr peak (514 keV).

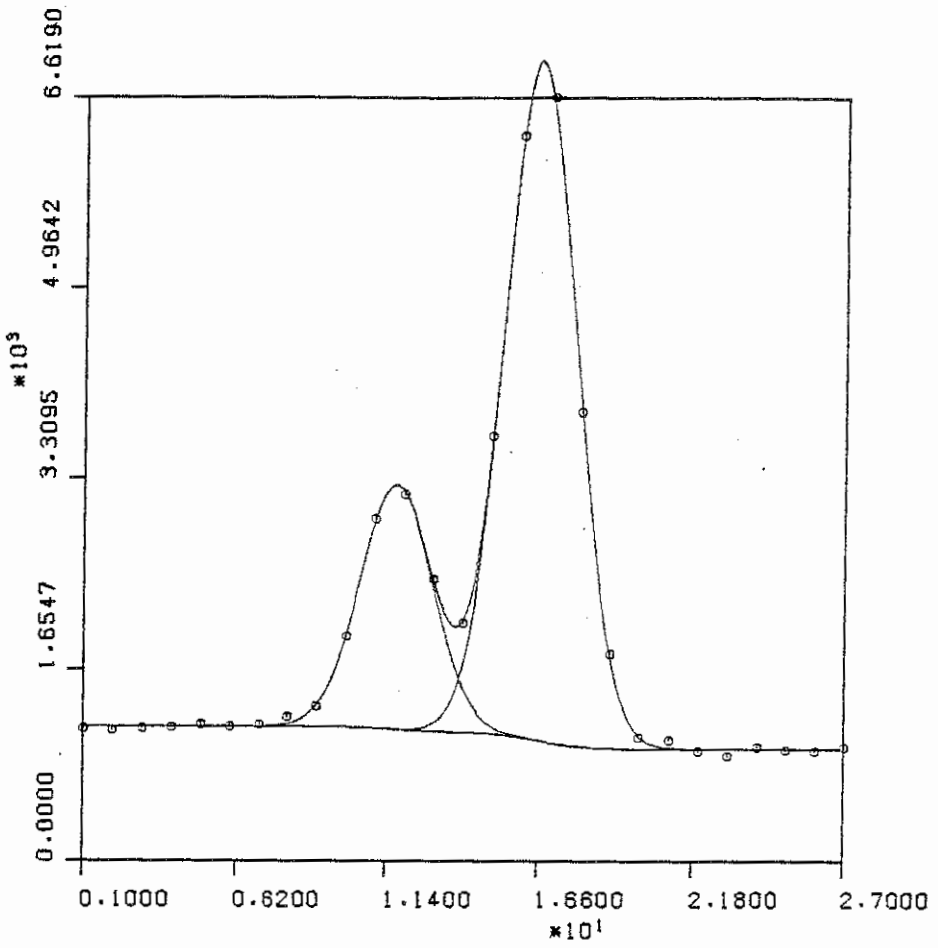


Fig. 4a. Example of peaks with relatively low Compton level.

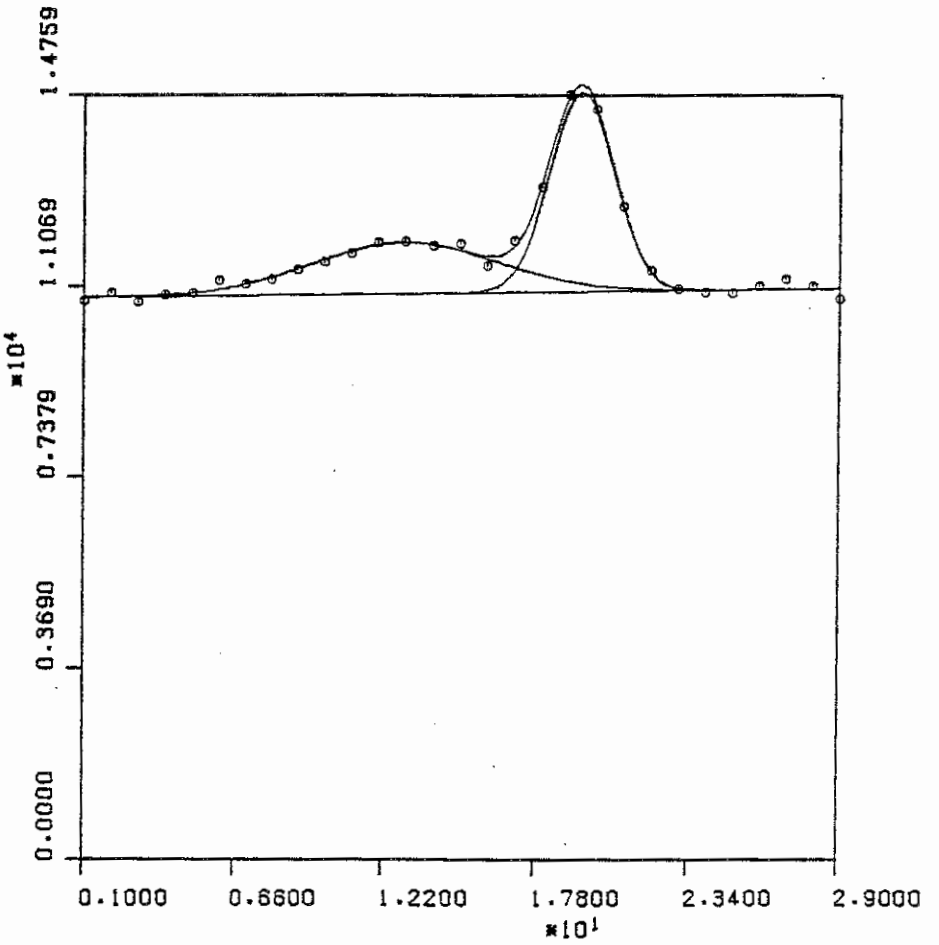
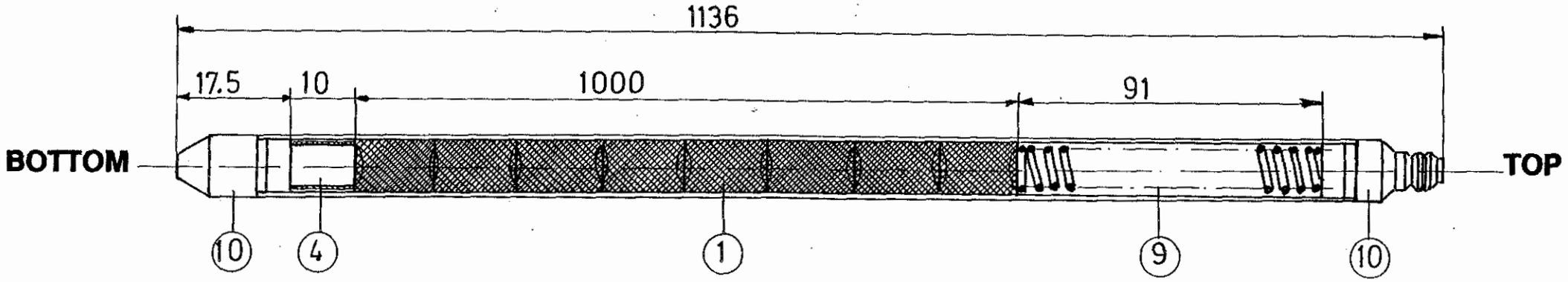
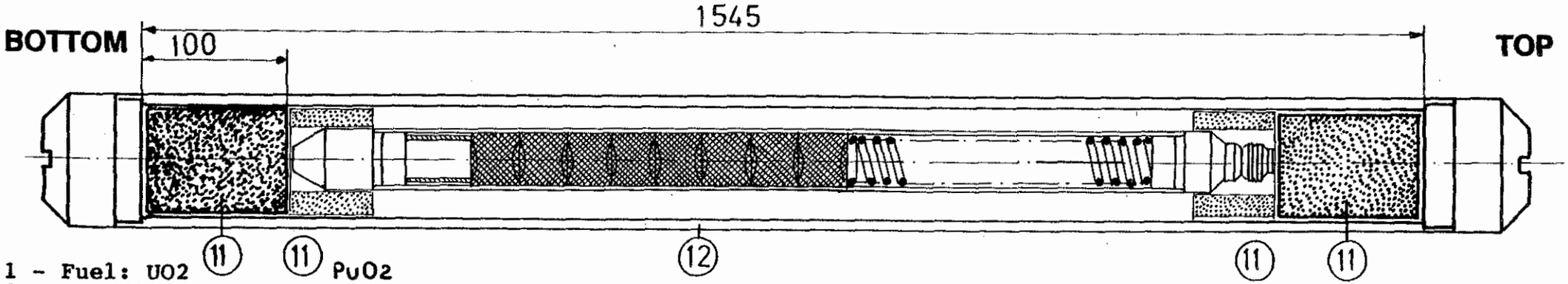


Fig. 4b. Example of peaks with relatively high Compton level.

A. DATA ON THE FUEL ROD



B. LOCATION OF A FUEL ROD IN A SCANNING TUBE



1 - Fuel: UO2
2 - Cladding: zircaloy4
3 - Intermediate grids of the assembly
4 - Pellet zircaloy4:
5 - Pellet Aisi 304:
6 - Pellet Natural UO2:
7 - Spacertube:
8 - Spacertube:
9 - Gasplenum with spring:
10 - Endstops of zircaloy4:
11 - Centerpieces of teflon:
12 - Scanning tube made of stainless steel(Φ27x33.5mm)

Fig. 5 : Schematic view of a fuel rod and of its location in a guiding tube for scanning