94/C6142/MBR/mvg/P-38

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STUDIECENTRUM VOOR KERNENERGIE
CENTRE D'ÉTUDE DE L'ÉNERGIE NUCLÉAIRE

DETERMINATION OF FISSILE MATERIAL BY NEUTRON TRANSPORT INTERROGATION:

Computer simulations of the neutron transport

Final Report 1992-1994

Contract FI2W-CT90-0010
coordinated by KFA Jülich

Michel Bruggeman and Robert Mandoki

Project leader: Pierre Van Iseghem

BLG-662

Mol (Belgium), September 1994

Work performed in the framework of the 1991-1995 programme of the European Atomic Energy Community: "Management and Storage of radioactive waste", Task 3: "Testing and evaluation of conditioned waste and engineered barriers"

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Abstract

This report summarizes the work performed at the SCK•CEN Mol in the period 92-03-15 till 94-03-15 in the framework of the CEC contract FI2W/0010. The research is done in collaboration with KFA Jülich, coordinator of the contract.
The research and results which are described in this report concern the evaluation and optimization by means of computer simulations of simple active and passive neutron systems for the assay of fissile material in waste packages. The starting point and impetus for this research is an existing active neutron-assay system at the KFA Jülich. This system uses a photon-neutron Sb-Be source to induce fissions in the fissile material present in the waste package. Prompt fission neutrons are detected and counted (total neutron counting). This simple neutron system has a detection limit below 1 g ^{235}U with a measuring time of 1000 s. The Sb-Be source, unfortunately, is impractical due to its high gamma output which needs severe gamma shielding and the rather short half-live of the source (60 days). The main purpose of this numerical research is to evaluate and optimize active neutron assay systems similar to the Sb-Be system but which uses more suitable neutron sources e.g. an Am-Li source. The assay systems described and studied in this report all have the common characteristic of being very simple. They only have one neutron detector tube and a neutron source fixed in space and time. The simplicity of these systems makes them especially attractive because:

- they are easy to use (no complex electronics, steady state neutronics);
- they are easy to manipulate and transport;
- their simplicity (neutron interrogation, detection, and counting) contribute to a highly reliable system;
- they are low cost systems.

The fact that the proposed systems are simple however does not exclude that their design and optimization still uses many aspects of neutron physics. The research performed at the SCK•CEN was entirely based on computer simulations of the neutron transport in the elements of the proposed apparatus and waste package. The one dimensional discrete ordinates code DTF-IV and the three dimensional Monte Carlo code MCNP were used to assess the different parameters important to the optimization and study of the neutron assay systems. A new system configuration with an Am-Li source is proposed for which a detection limit of 10 g ^{235}U was calculated. Although this detection limit is an order of magnitude higher than the one of the Sb-Be neutron system it is believed that the performances of this system still can be improved. For an active neutron system completely identical to the Sb-Be system, but with a Am-Li source, it is shown that the signal to noise ratio of the counter can be optimized using tayloring of the neutron-energy spectrum and efficient energy selective neutron transport. It is also shown that in the passive counting mode the neutron counter has a detection limit of 30 mg plutonium for total neutron counting.

Objective and scope of contract FI2W-CT90-0010

This research was concerned with nondestructive assay techniques for fissile material determination in waste material mainly in waste drums. The starting point for development work was an assay system at the KFA for fissile material determination by active neutron interrogation with an Sb-Be neutron source. In this assay system the fission neutrons were discriminated from the source neutrons by their transport properties in hydrogenous material. The neutron count rate was composed of a source term and a second term proportional to the fissile material content of the investigated sample. The system directly determines all the fissionable nuclides U-233, U-235, Pu-239 and Pu-241. It required shieldings due to the approximately 2E12 Bq Sb-124 and had detection limits between 1 mg and 1 g fissionable material depending on sample size and matrix composition.

Neutron transport calculations at SCK•CEN Mol were intended to achieve a theoretical understanding and an improvement of the assay system with different neutron sources by modelling the neutron transport properties in the waste drum and the assay system. A replacement of the Sb-Be neutron source by other low energy neutron sources, such as Am-Li, will lead to an assay system with minimum shielding requirements and constant source strength (Am-241, $t_{1/2}$ = 432.6 y, Sb-124, $t_{1/2}$ = 60.3 d). As an additional advantage there is no need for a reactor to reactivate the Sb. The replacement of the Sb-Be neutron source by Am-Li was therefore an important objective of this research.

Passive neutron emission mainly results from spontaneous fission in Pu-238, Pu-240, Cm and Cf isotopes. Counting these neutrons gives information on the presence of these transuranic elements in the waste matrix. Effective recording and evaluating of these neutrons is an objective of this research. The aim of the final assay system is an easy-to-use and reliable instrument for the estimation of determination of the fissile material content of various packages, mainly waste drums.

Work programme of contract FI2W-CT90-0010

1) Checking and optimization of the Sb-Be system by comparison with neutron transport calculations.
2) Active neutron interrogation with other neutron sources, in particular Am-Li and comparison with neutron transport calculations.
3) Modification of the system for passive neutron counting capabilities.
4) Test and performance of the active/passive neutron assay system with actual samples, mainly drums with waste from the nuclear fuel cycle.

This report summarizes the contributions of the SCK•CEN to the contract FI2W-CT90-0010. Detailed descriptions of the progress of the work can be found in the earlier progress reports [Mandoki-92, Mandoki-93, Mandoki-94]

Contents

A Introduction

Neutron interrogation is a nondestructive assay technique used to measure the mass of fissionable material ^{235}U, ^{239}Pu, ^{241}Pu... in waste packages. The technique is based on the detection of the neutrons originating from induced fission reactions in the fissionable material. The fission reactions are induced by neutrons emitted by an intense external source. The neutron flux necessary to interrogate the waste package is generally orders of magnitude higher than the resulting neutron flux detectable from the induced fissions. As a consequence the small variation of the total neutron flux due to the fissioning material in the interrogated waste package is not easily measured or detected.

Neutron detectors generally cannot discriminate between the source neutrons and the fission neutrons and an additional discrimination process is needed to prevent the source neutrons of being detected or to reduce the background of source neutrons. The discrimination between the two origins of neutrons generally is accomplished using pulsed neutron sources or isotopic neutron sources shuffled between an irradiation position and a shielded position. The neutrons originating from the fission reactions are then measured when the source is switched off or removed. These neutron interrogation techniques need expensive neutron generators, complex counting electronics and electro-mechanics in the case of a shuffled source.

A less commonly used neutron interrogation technique for waste assay uses an interrogation source fixed in space and time (f.i. the source is not withdrawn or switched on and off). This technique is successfully applied in active neutron collars used for accountability measurements in safeguards applications where relatively high masses of fissionable material are measured in relatively small containers. The extension of this technique to waste packages (e.g. 200 l drums with a concrete waste form matrix) containing only small quantities of fissionable material is not evident.

P. Filß from the KFA Jülich demonstrated the applicability of a neutron-interrogation waste assay system using an Sb-Be photon-neutron source which is continuously irradiating the waste package. A typical detection limit of 1 g ^{235}U was obtained with this system [Filß-90]. The main advantages of this assay system are its simplicity and the ease to transport the system and carry out the measurements at another site. However there is a major problem connected to the use of an Sb-Be source. Although this source has a very favourable neutron energy spectrum for the proposed application, it needs severe gamma shielding and the source has a rather limited operational period due to the half-life of only 60 days. These were the main reasons for further investigating this particular neutron interrogation technique but with other, more practical neutron sources. Unfortunately most sources have a less favourable neutron-energy spectrum and an optimization of this interrogation technique logically needs further improvements to obtain comparable detection limits as with the Sb-Be-neutron system.

In the following paragraphs the principles of time- and energy-discrimination are first discussed because of their importance in the understanding of the active neutron assay system and related problems dealt with further in this document.

Time-discrimination

Commonly used neutron interrogation techniques apply time-discrimination to distinguish between the neutrons from the external source and the neutrons from the fission reactions. Using time-discrimination, induced fission neutrons are only measured when the external source is switched off or removed to a shielded position. The removal or the switching of the source is done very fast. When the source is off or withdrawn, the flux of source neutrons does not drop immediately to zero but decays to zero. The decay constant depends on the characteristics of the constituents of the waste package and its surroundings. The decay of the fast-neutron flux is dominated by the moderation process which transforms high-energy neutrons in to low-energy neutrons. The low-energy neutrons induce fission reactions or are absorbed in the waste matrix and its surroundings. Time discrimination is used in two well-known techniques: the differential die-a-way technique (DDT) and the californium shuffler. With the differential die-a-way technique the prompt neutrons from the induced fission reactions are measured after high-energy neutrons of an intense neutron burst have thermalised and irradiated the waste package. In a similar way the shuffler technique uses a mechanical way to transport (shuffle) an isotopic source (mostly ^{252}Cf) forth and back between an irradiation position and a shielding. Delayed fission neutrons are measured after the source is withdrawn into the shielding.

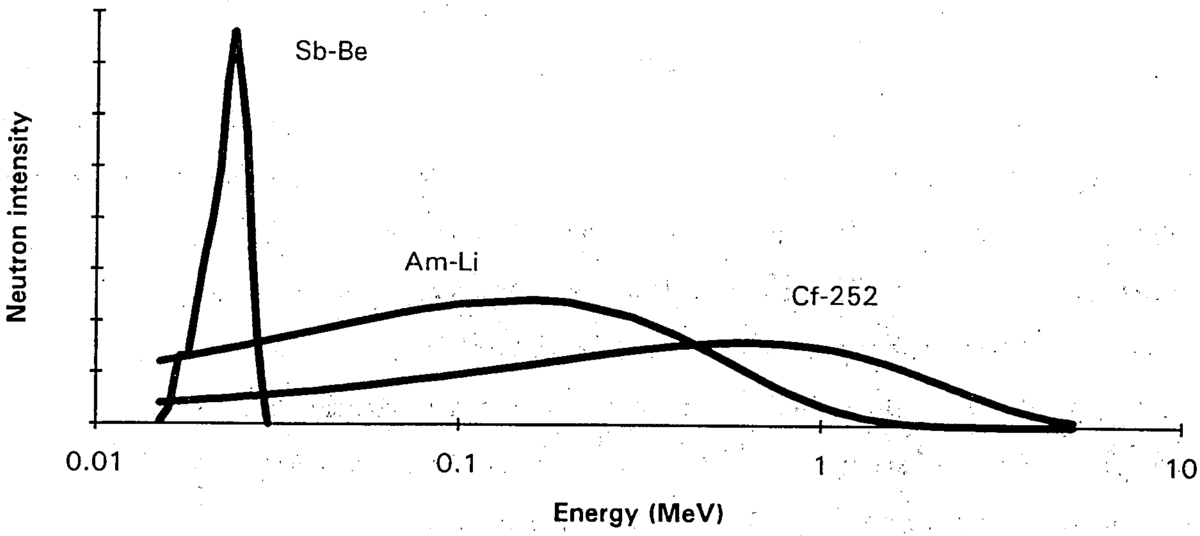

Figure 1 Neutron-energy spectrum for A) Sb-Be, B) Am-Li, C) fission spectrum Cf-252.

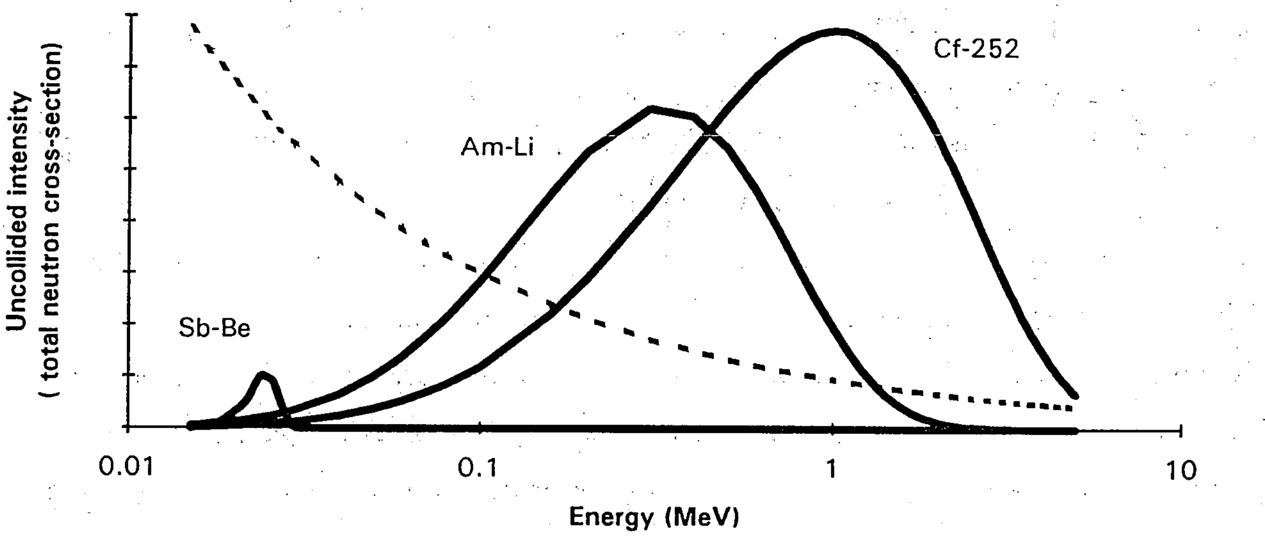

Figure 2 Evolution of the total neutron -interaction cross-section (dotted curve,$E^{-1/2}$ evolution) and uncollided neutron intensities (see text).

Energy-discrimination

A less commonly applied discrimination technique used in the neutron counting in active waste assay systems is based on energy-discrimination between source neutrons and fission neutrons. This discrimination method was demonstrated by P. Filß using a very simple assay system for the determination of fissile material in 220 l waste drums. In this system a neutron source emitting neutrons with a low mean energy is at a fixed position with respect to the waste package and is continuously irradiating it with neutrons. The energy-discrimination is based on a energy-selective neutron transport of high- and low-energy neutrons through material. High-energy neutrons will penetrate deeper in the material while low-energy neutrons will suffer absorption resulting in poor penetration. The selective transport combined with the detection of neutrons can be designed to detect preferentially fast neutrons and to have low sensitivity for low-energy neutrons. An ideal energy discrimination clearly is obtained applying low-energy neutrons for the interrogating source, and detecting the high-energy neutrons from the induced fission events (explaining why a Sb-Be source was used which has a low mean neutron energy). The interrogation with thermal or near-thermal neutrons also takes advantage of the large fission cross section in the fissile material (about 1000 barn) for fission by thermal or near-thermal neutrons. The fission by fast neutrons (10 keV to 10 MeV) has a fission cross section of only 1 to 2 barn. All elements explaining and visualizing the whole concept of the energy discrimination are summarized in figures 1 and 2. Figure 1 shows the energy spectra of two isotopic sources with a low average neutron energy. Spectrum A shows the spectrum of the Sb-Be photon-neutron source which has an average neutron energy of 24 keV, spectrum B corresponds to the neutron-energy spectrum of an Am-Li source which has a average neutron energy of 500 keV. The spectra of these isotopic sources, which are candidate sources for a good energy discrimination, have to be compared with the mean neutron energy emitted in an induced fission reaction which is approximately 2.3 MeV. A typical fission neutron-energy spectrum is shown on figure 1 (spectrum C). The properties of the neutron interaction with material and the efficiency of the energy selective transport of low- and high-energy neutrons are visualized in figure 2 showing the evolution of the total neutron-interaction cross-section as a function of the neutron energy σ_t (E). A $1/\sqrt{E}$ dependence was supposed here for simplicity (dotted curve on figure 2). The cross-section decreases with increasing energy making the material more transparent for high-energy neutrons than for low-energy neutrons. Figure 2 also shows the uncollided intensities $I(E,x) = I_0(E) \exp(-N \sigma_t (E) x)$ for the different spectrum components. In this expression $I(E,x)$ is the uncollided neutron intensity after transport through a layer of thickness x and atom density N. $I_0(E)$ is the intensity before entering the layer. The uncollided intensity is a measure for the efficiency of the selective transport. Figure 2 shows that Sb-Be spectrum is strongly attenuated while the fission spectrum is much less attenuated. The selective transport acts as a high-pass filter for high-energy neutrons.

From the figures 1 and 2 the main problems to deal with in the optimization of the selective transport may already be identified:

a) although the considered isotopic neutron sources have a mean neutron energy well below the mean energy of the fission neutrons, their neutron-energy spectrum always shows a more or less pronounced high-energy tail;

b) the neutron interaction cross-section does not show a sharp transition from high energy to low energy and as a consequence energy selective transport through matter will never act as a perfect high-pass filter for fission neutrons.

The implementation and optimization of neutron energy-discrimination through selective transport needs knowledge and insight in the neutron physics of the interaction of neutrons with matter. Qualitative and quantitative aspects are most of the time only assessable through the experiment or via computer simulations. In an initial phase computer simulations are to be preferred because they offer the possibility of an easy choice of materials, geometries, source spectra... .

The following paragraphs of this report will mainly deal with the computer simulations related to:

- choice and optimisation of the geometrical configuration of detector and interrogation source;
- influence of the source spectrum: Sb-Be, Am-Li, ^{252}Cf, accelerator spectrum;
- evaluation of materials leading to an optimal selective neutron transport and detection;
- shielding of the source neutrons in the direction of the detector;
- distribution of interrogating neutron flux in the waste package;
- study of the performances of the system when used as a passive neutron counter.

The active and passive assay systems which will be described and discussed further in this text use only one neutron detector tube which might be a ^{3}He or BF_3 detector. These detector tubes respond essentially to low-energy neutrons. If this detector tube is imbedded in a moderator such as polyethylene (PE) the combined system will principally respond to fast neutrons with a high detection efficiency (better than the detection efficiency of a ^{4}He neutron detector). The detector block (PE + detector tube) is said to be a fast-neutron detector. Thermal neutrons entering the PE detector block normally are absorbed in the PE before they can interact with the detector volume. The neutron-interrogation system which we will consider has an isotopic neutron source which is fixed close to the waste package.

B Geometrical configuration of the active neutron assay system

The geometrical configuration of the active system describes the way the two main components (the detection unit and the external source) are positioned with respect to the waste package. Although several arrangements could be considered we will concentrate only on two different and well-defined configurations which are described and shown below in the figure 3 a and b).

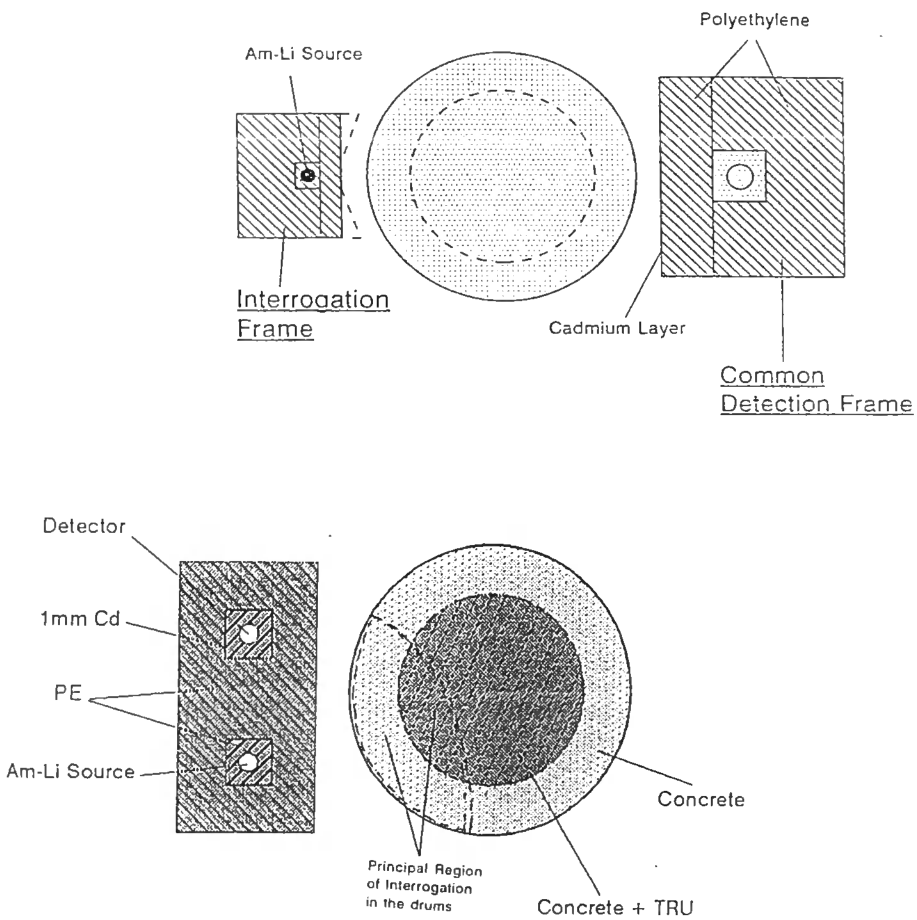

Figure 3 Geometrical configurations of the active neutron system.
a) Source-Barrel-Detector configuration (SBD)
b) Detector-Source-Barrel configuration (DSB)

1) The configuration in which the neutron source, the waste drum and the detector block are on a straight line with the waste drum positioned between the source and the detector block will be referred to as the SBD-layout because of the order **S**ource-**B**arrel-**D**etector (see fig. 3 a).
2) The configuration in which the detector block and the neutron source are both at the same side of the drum will be referred to as the DSB-layout (**D**etector-**S**ource-**B**arrel) (see fig. 3 b).

The original configuration demonstrated by P. Filß was a SBD-configuration, however it was believed that a DSB configuration could improve the detector response to neutrons of the induced fissions in the fissile material. Arguments in favour of a DSB configuration take into account that the low-energy neutrons of the interrogation source have a poor penetration depth (short mean free-path-length) in the drum and as a consequence the induced fissions principally will occur in the drum-half close to the source. With a DSB detection configuration fission neutrons emitted in the drum-half close to the source have to travel less distance before they can be detected. This clearly will result in a higher detection efficiency. Concerning the DSB configuration there is a major drawback related to the fact that the source is close to the detector; this will undoubtedly result in a high neutron background. This configuration clearly will need a very efficient shielding between the detector block and the neutron source without loss in detection efficiency for the fission neutrons. In the SBD layout this shielding is less important because the waste drum itself acts as a shielding. The advantages of one configuration or another have to be evaluated experimentally or via computer simulations of the neutron transport.

In the numerical evaluations of both configurations the following assumptions concerning the detection and neutron source were made:

A) The neutron detector block contains only one ^{3}He-detector tube imbedded in a PE block (possibly wrapped in a thin cadmium liner). To distinguish between a bare ^{3}He detector and the detecting assembly of detector tube + PE we will refer to the latter as detector block.
B) The neutron sources that will be considered are isotopic neutron sources such as Am-Li, Sb-Be, ^{252}Cf .... The neutron spectrum of these sources however may be modified by the use of

a moderator slab. This moderator has a filter-function transforming the high-energy neutrons into low-energy neutrons.

C Shielding between the Am-Li source and the detector block

As was explained in the previous paragraph the DSB-configuration of the active neutron waste assay system needs an efficient shielding between the neutron source and the detector block to reduce the source background at the detector tube. The main aim of the computer simulations described in this paragraph is to investigate the shielding of the Am-Li source neutrons in the direction of the detector tube in order to arrive at an acceptable neutron background level. The candidate shielding materials that we considered are:

iron, aluminium, graphite, PE and PE combined with a thin Cd layer.

The shielding problem was investigated numerically using the one dimensional discrete ordinate neutron transport code DTF-IV. In all calculations the distance (centre to centre) between the Am-Li-source and the detector tube was kept constant at 25 cm . The dimensions of the source and the detector along the one-dimensional axis was 4 cm (see figures 4 and 5). Respective layers of 5 and 10 cm of the shielding materials were considered. A PE-layer of 3 cm was always placed between the source and the shielding material. The residual space in front of the detector tube was always filled-up with PE. In all calculations the neutron flux at the centre of the source was defined to be 1 n/s cm^2. The spacial ordering of the materials between source and detector tube and their respective dimensions are given in table 1.
For all cases that were studied the total, fast and thermal neutron fluxes were calculated as a function of the position between source and detector. Only two typical graphical results are shown here in figures 4 and 5. They show the evolution of the neutron fluxes as a function of the position in the source-detector-block. Figure 4 shows the results for a PE shielding combined with a cadmium lined PE detector block. Figure 5 shows the results for a PE-Fe-PE shielding. The calculated neutron fluxes at the position of the detector tube for all the considered cases are summarized in Table 2. The optimal source shielding corresponds to the case resulting in the lowest thermal neutron flux ($E < 0.4$ eV) at the site of the detector tube. From Table 2 we see that this corresponds to case 4, where the shielding is composed of PE and a Cd layer (1 mm). In all

cases the fast neutron flux (E > 0.1 MeV) is approximately 1 order of magnitude smaller than the thermal flux.

Case	Shielding material and spacial ordering	Layer thickness (cm)
1	PE	21
2	PE Al PE	3 5 13
3	PE Fe PE	3 5 13
4	PE Cd PE	17.9 1 mm 3
5	PE Fe PE	3 10 8
6	PE Al PE	3 10 8
7	PE graphite PE	3 10 8

Table 1 Shielding materials and their spacial ordering going from the source to the detector tube.

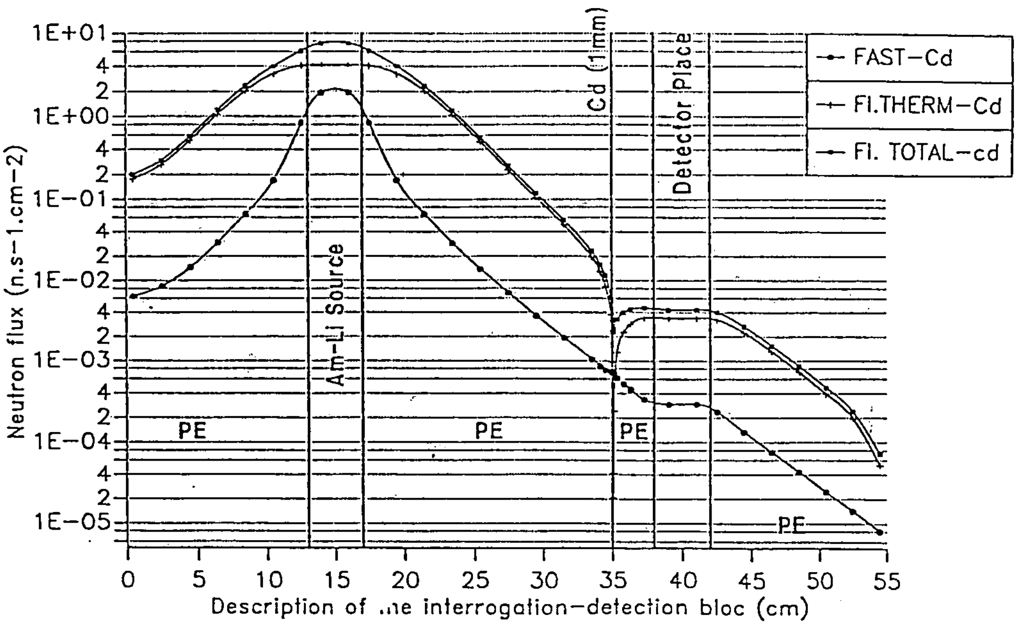

Figure 4 Neutron fluxes in the source-detector-block calculated for case 4 (PE-Cd-PE).

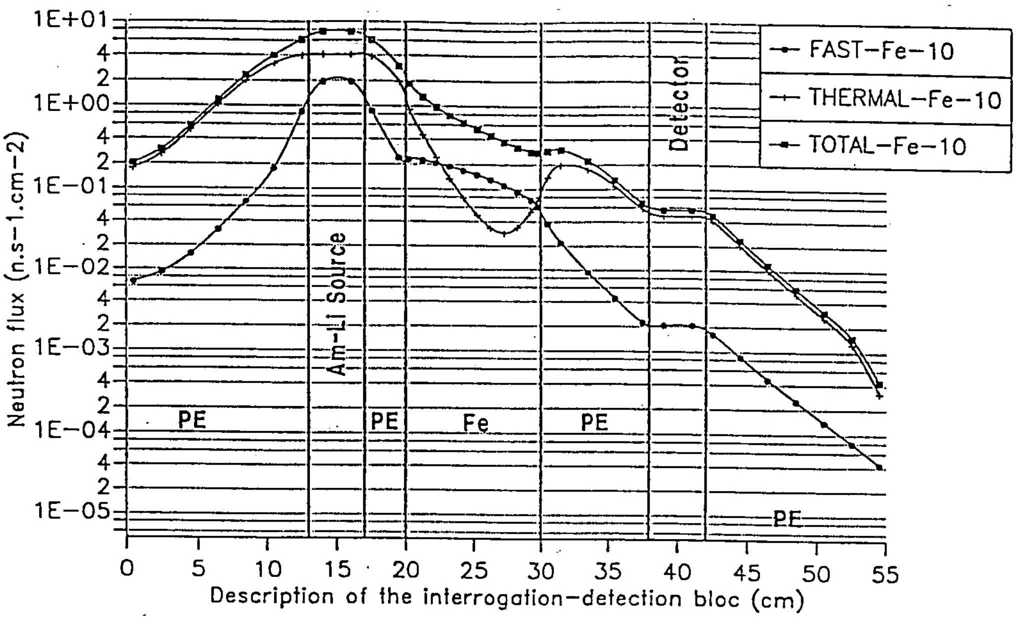

Figure 5 Evolution of the calculated neutron fluxes in the source-detector-block (case 5 PE-Fe-PE)

Case	1	2	3	4	5	6	7
Detector position							
Total flux	8.3 E-04	2.4 E-03	2.2 e-03	5.8 E-04	7.3 E-03	1.0 E-02	3.4 E-03
Thermal flux	7.1 E-04	2.1 E-03	1.8 e-03	4.5 E-04	6.3 E-03	9.0 E-03	3.0 E-03
Fast flux	3.8 E-05	1.0 E-04	1.0 e-04	3.9 E-05	2.8 E-04	2.8 E-04	8.4 E-05
Flux attenuation							
Total flux	1203	409	463	1728	136	98	298
Thermal flux	774	259	297	1208	86	58	197
Fast flux	6657	2533	2581	6526	935.8	941	2753

Table 2 Total, thermal and fast neutron fluxes (n cm^{-2} s^{-1}, Am-Li source 1 n s^{-1} cm^{-2}) at the detector-tube position. The last three rows give the flux attenuations for the respective neutron fluxes. The different cases are described in Table 1 and [Mandoki-92].

Although it is well known that high-density PE is a good shielding for neutron sources and is commonly used for radiation-protection applications it was believed that combinations of PE with iron, aluminium or graphite possibly could improve the shielding of essentially the thermal neutron flux at the position of the detector tube. Table 2 clearly proves that this is not true and that PE combined with a Cd-layer is the best choice. Of course this shielding could still be improved adding efficient absorbers of thermal neutrons near the detector tube. However we should keep in mind that the absorption of thermal neutrons near the detector will also decrease the detection efficiency for fission neutrons. Further improvement may be obtained if the distance between the source and detector is increased but increasing this distance will also decrease the interrogation flux in the waste barrel. The main conclusion from these calculations is that the combination PE and Cd slab used as shown in Figure 4 gives the best shielding results.

D Modification of the energy spectrum of the Am-Li source

The mean neutron energy of the neutrons from the Am-Li source is appreciably higher than that of the Sb-Be source. Using moderating materials the spectrum of the neutron source can be *softened.* This action on the energy spectrum is generally referred to as tayloring of the source spectrum. The tayloring of the neutron-energy spectrum of the Am-Li interrogation source aims at optimizing this spectrum, to maximize the ratio of detected fission neutrons to detected source neutrons N_f/N_s. This quantity is proportional to the ratio $\Delta\Phi/\Phi_s$ in which $\Delta\Phi$ (the signal) is the variation in neutron flux with the variation in mass of fissile material present in the waste package and Φ_s (the source background) the flux of source neutrons. The signal to noise ratio $\Delta\Phi/\Phi_s$ mainly depends on:1994-11-16

- the position of the detector with respect to the source (SBD or DSB configurations);
- the number of induced fissions in the waste drum;
- the selective transport (attenuation) of the flux of source neutrons;
- characteristics of the matrix of the waste package;
- detector characteristics.

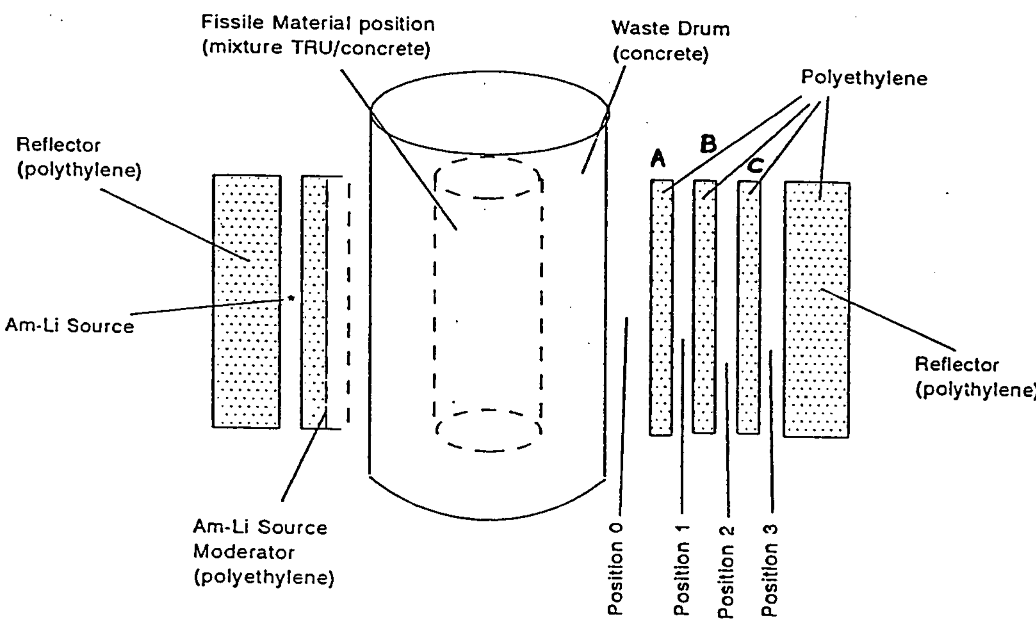

Figure 6 Input geometry for the calculation of neutron fluxes.

The number of induced fissions at a particular position in the waste drum is proportional to the thermal neutron flux at that position. The thermal flux distributions in the drum can, to a certain degree, be modified using moderating slabs between the waste drum and the neutron source. Moderation will *soften* the energy spectrum giving more weight to the low-energy components in the spectrum. Spectrum *hardening* is in contradiction with the proposed idea of energy-discrimination. The influence of a moderating slab of PE on the fast and thermal neutrons was evaluated at the source-side of the drum and at the rear-side (seen from the source position).

These positions correspond to the detector-tube positions of the SBD and DSB configuration respectively. For the SBD layout the influence of the thickness of a PE slab between detector tube and waste drum was also considered. In all cases the impact of the PE-slabs acting as a selective transport medium were evaluated considering the ratio $\Delta\Phi/\Phi_s$ for a mass of 10 g ^{235}U distributed homogenously in the inner region of the 220 l waste drum (the drum was thought to have 10 cm thick concrete walls). The input geometry used for this series of calculations with the DTF-IV code is sketched in Figure 6. The thickness of the PE slab between the Am-Li source and the drum was varied between 0 and 15 cm in steps of 5 cm. The PE slabs A, B and C have each a thickness of 5 cm and are stacked horizontally with a spacing corresponding to the detector diameter. The quantity $\Delta\Phi/\Phi_s$ was evaluated at Position 0 up to Position 3 corresponding to 4 different detector positions determined by the PE slabs (see Fig. 6). The results of these calculations are summarized in Table 3.

From this table the following conclusions can be drawn:

- The mean rate of induced fissions in the waste drum significantly decreases when the source spectrum is moderated with a PE slab (see second row of Table 3);

- The ratios $\Delta\Phi/\Phi_s$ evaluated at a position between the source and the drum do not show any significant variation with the presence of fissile material in the waste package;

- At the rear side of the drum (SBD positions) both quantities $\Delta\Phi/\Phi_s$ (therm and fast) are sensitive to the presence of fissile material;

- The optimum of the ratio $(\Delta\Phi/\Phi_s)$therm is obtained for a 5 cm thick PE layer between the Am-Li source and the drum;

- The ratios $\Delta\Phi/\Phi_s$ increase with increasing thickness of the PE-layer between detector and drum. The discrimination is better with increasing thickness of the PE slab in front of the detector tube.

PE thickness between source and drum (cm)		0	5	10	15
Fission rate (E-3) (1/s)*		38.3	9.8	1.7	0.3
Position at source side	$(\Delta\Phi/\Phi_s)_{therm}$	/	0.1	0.2	0.2
	$(\Delta\Phi/\Phi_s)_{fast}$	0.0	0.0	0.0	0.0
Position 0	$(\Delta\Phi/\Phi_s)_{therm}$	1.2	1.8	1.7	1.4
	$(\Delta\Phi/\Phi_s)_{fast}$	86.8	123.1	85.9	56.5
Position 1	$(\Delta\Phi/\Phi_s)_{therm}$	7.4	10.2	7.6	5.9
	$(\Delta\Phi/\Phi_s)_{fast}$	143.2	191.2	129.7	83.8
Position 2	$(\Delta\Phi/\Phi_s)_{therm}$	33.3	48.0	27.6	19.3
	$(\Delta\Phi/\Phi_s)_{fast}$	183.5	239.3	160.8	103.4
Position 3	$(\Delta\Phi/\Phi_s)_{therm}$	80.4	118.4	56.5	36.4
	$(\Delta\Phi/\Phi_s)_{fast}$	228.4	292.6	509.7	125.2

Table 3 Signal to noise ratios $(\Delta\Phi/\Phi_s)$ (for a mass of 10 g ^{235}U) evaluated at different thicknesses of the PE-moderator (columns) and for different thicknesses of the PE selective transport layer (position 0... position 3). * *(for an initial source strength of 1 n s^{-1} cm^{-2})*

Results of the calculations (not presented here; see [Mandoki-93]) show also that the neutron fluxes Φ_f of fission neutrons and Φ_s of source neutrons both strongly decrease with increasing thickness of the PE discrimination layer between the drum and the detector tube (SBD configuration) [Mandoki-92]. In experimental situations the variance of the total number of detected counts has also to be considered for a certain defined measuring time and intensity of the external source. These factors will determine the statistical precision of the measurement. Considering experimental factors the optimum will not be obtained with the thickest PE layer.

There is no observable variation in the neutron fluxes on the source-side of the waste drum when fissile material is present in the drum. We should remember that in that particular position the source intensity is still very high and relative small variations in the neutron fluxes cannot be observed at that particular position. The effect of the different PE layers can be summarised as follow:

- A PE layer between source and the waste package attenuates the thermal- and fast- neutron fluxes. The attenuation, although a complete energy spectrum of neutron energies is present, can be described quite well with a mean-free-path length λ valid for the whole neutron-energy spectrum. The mean-free-path length in PE was evaluated to be 2.86 cm;
- A PE layer between waste package and detector tube also attenuates the neutron fluxes (from the source and from the induced fissions) but no common mean-free-path lengths can be deduced for thermal- and fast-neutron fluxes. Table 4 gives the calculated mean-free-path lengths for thermal and fast fluxes of respectively the source neutrons and for a composed spectrum of source neutrons and neutrons from the induced fission reactions (10 g ^{235}U). The reported values should be interpreted with care e.g. λ_{therm} and λ_{fast} may not really be considered as mean-free-path lengths because they are defined for energy intervals and not for a single energy. The constants λ_{therm} and λ_{fast} are larger for the combined spectrum than for the source spectrum alone. This clearly illustrates the effects of the selective transport.

In a second series of calculations we investigated the performances of graphite as a selective transport medium [Mandoki-92]. We evaluated the effect of using graphite slabs instead of PE slabs as a selective transport medium in combination with the detector tube. Graphite has a higher moderating ratio which is approximately 200 compared to 122 for PE. Due to this, a graphite moderator in combination with a ^{3}He or BF_3 detector tube will yield a higher efficiency for the detection of fast neutrons.

Mean-free-path lengths in the PE layers between drum and detector tube	λ_{therm} (cm)	λ_{fast} (cm)
Am-Li source spectrum + fission spectrum*	3.03	4.00
Am-Li source spectrum (background)	1.82	3.13

Table 4 Mean-free-path lengths in PE. The spectrum descriptions given in the first column of this table are for the neutron spectra influenced (moderation and absorbtion) by the waste package! (*) Values for 10 g ^{235}U.

To evaluate graphite as a selective transport medium in this specific problem, we repeated the calculations described in the previous paragraph but with the PE slabs A, B and C replaced by graphite slabs (see Figure 6). The results of these neutron calculations are summarized in Table 5. Table 5 gives the results for the case of a 0 respectively a 5 cm thick PE slab placed between the drum and the Am-Li source for spectrum tayloring. In both cases the ratios $\Delta\Phi/\Phi_s$ are considered in Positions 0 to 3 in a similar way as with the PE slabs. The quantities $\Delta\Phi$ (therm and fast) were calculated for 10 g ^{235}U. Table 5 should be compared with Table 3. The comparison shows that a graphite moderator clearly gives smaller values for the ratios $\Delta\Phi/\Phi_s$ and the discrimination effect is not better for materials with a higher moderating ratio. For this particular application graphite clearly does not improve the selective transport. However a combination in which a PE slab for selective transport is combined with graphite as the moderator material of the detector bank (possibly wrapped in a Cd-layer) might give a further improvement compared to the case where PE is used for both applications.

PE thickness (cm)		0	5
Position 0	$(\Delta\Phi/\Phi_s)_{therm}$	1.3	3.3
	$(\Delta\Phi/\Phi s)_{fast}$	193.5	113.9
Position 1	$(\Delta\Phi/\Phi_s)_{therm}$	1.7	4.8
	$(\Delta\Phi/\Phi_s)_{fast}$	106.3	145.2
Position 2	$(\Delta\Phi/\Phi_s)_{therm}$	2.2	6.9
	$(\Delta\Phi/\Phi_s)_{fast}$	135.1	179.0
Position 3	$(\Delta\Phi/\Phi_s)_{therm}$	2.7	9.6
	$(\Delta\Phi/\Phi_s)_{fast}$	165.8	212.2

Table 5 Signal to noise ratios $(\Delta\Phi/\Phi_s)$ (for a mass of 10 g ^{235}U) evaluated at different thicknesses of the PE-moderator (columns) and for different thicknesses of a graphite selective transport layer (position 0... position 3, see Figure 6).

E Neutron flux distribution in the waste drum

A more thorough study of the flux distributions of the source neutrons in the waste drum was initiated using the Monte Carlo code MCNP-4.2 [Mandoki-93]. The knowledge of the three dimensional repartition of the interrogating neutron flux in the waste drum is interesting as it will give information about: the way the fissile material is interrogated, about systematic assay errors due to the non uniform interrogation and the neutron flux in each part of the drum.

To perform these calculations the drum with a concrete waste form matrix ($d = 2$ g/cm^3) was divided in 44 × 7 cells sectioning the drum in 7 segments along the drum axis and subdividing each segment in 44 cells as shown in Figure 7. The thermal- and fast-neutron fluxes were calculated in each cell. Two different neutron-energy spectra were considered: the Am-Li spectrum and the ^{252}Cf spectrum. The ^{252}Cf source spectrum was considered here because of its average energy of 2.14 MeV which will result in deep-penetrating neutrons. Calculations were performed for each of these sources positioned at different locations with respect to the drum and to a PE block in which they were thought to be imbedded. The considered positions are indicated in Figure 7 and are referred to as P1 up to P6. The positions P1, P2, P3 and P4 correspond to the situation in which the source is respectively separated from the drum by 0, 1, 2 and 3 cm of PE. The positions P5 and P6 have different meanings for the two sources: for the Am-Li source P5 and P6 correspond to 1 cm of PE seen along the direction of the x-axis, for the ^{252}Cf source P5 and P6 correspond to 3 cm of PE along this direction.

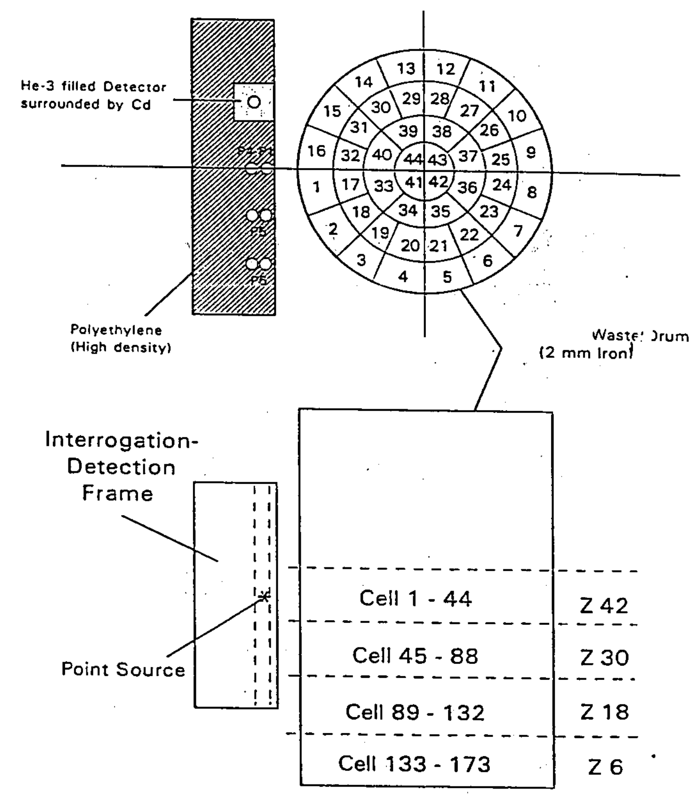

Figure 7 Cell meshing used in a section and along the drum axis to calculate the 2D-neutron-flux distributions in the waste package.

Typical results for the distribution of the thermal flux in the central segment of the drum (Z = 42 cm) and depending on the source positions P1, ... P6 are shown in Figures 8 and 9 for respectively the ^{252}Cf and Am-Li source. The flux distributions are visualized with equiflux lines. Variation of the equiflux lines as a function of the considered segment of the drum (only segments at 6cm, 18 cm, 30 cm and 42 cm were considered) are shown in Figure 10 for the Am-Li source. In all these figures the value of the flux (n/s cm^2) is given on the equiflux lines. The smooth curvature of the flux lines —seen the rather rough cell meshing which was used for the calculation— was obtained by fitting cubic-spline functions through the original data points.

Evaluating the Figures 8, 9 and 10 we can conclude that:

- The equiflux lines visualizing the thermal flux in the central segment of the waste package (Z=42 cm) may — in a good approximation — be considered as bow segments with a common centre (origin) at the source position. The same conclusion holds for the equiflux lines for the segments at positions Z = 6, 18 and 30 cm but here the centre point has moved towards the centre of the drum. This might be an interesting feature for an analytical modelling of the flux distribution;

- When the source is positioned in positions 5 or 6, then the flux distributions reoriented correspondingly and regions with lower thermal flux move in front of the detector. This will undoubtedly result in a lower detector response to fissile material, but on the other hand will decrease the source background.

- The effect of the spectrum moderation by the PE slab for both neutron sources clearly illustrates the deeper penetration of the neutrons from the ^{252}Cf source compared to the Am-Li source (compare positions P2..P4, remember that the positions P5 and P6 correspond to different thicknesses of the PE layer). There is no gain in using the ^{252}Cf source in the DSB configuration. Indeed for an unmoderated spectrum (no PE between source and drum) both sources give approximately equal thermal-flux distributions (Compare the positions P1 for both sources).

Considering the above conclusions and observations we should realise that a lot of information concerning the variation of the specific energy spectrum of the neutrons when they travel through different materials is finaly evaluated in the calculations only for two energy intervals indicated as fast and thermal. This is a rather poor sampling of the spectrum and probably interesting information that might be useful to gain more insight in the problems could be lost! The situation probably would also alter if neutron reflectors were put behind the drum.

The variation of the thermal flux along the axis connecting the source with the drum centre is shown in figures 11 and 12 for respectively the ^{252}Cf and Am-Li source. The different curves on each graph represent the thermal flux for the source in positions P1 to P4. The fact that the curves are straight lines on a logarithmic plot and are nearly parallel states that a common mean-free-path length — independent of the spectrum tayloring — can be defined to describe the variation of the thermal neutron flux in the waste package. For ^{252}Cf a mean-free-path length $\lambda_{Cf252} = 11.1$ cm in the concrete matrix was calculated while for Am-Li $\lambda_{AmLi} = 9.09$.

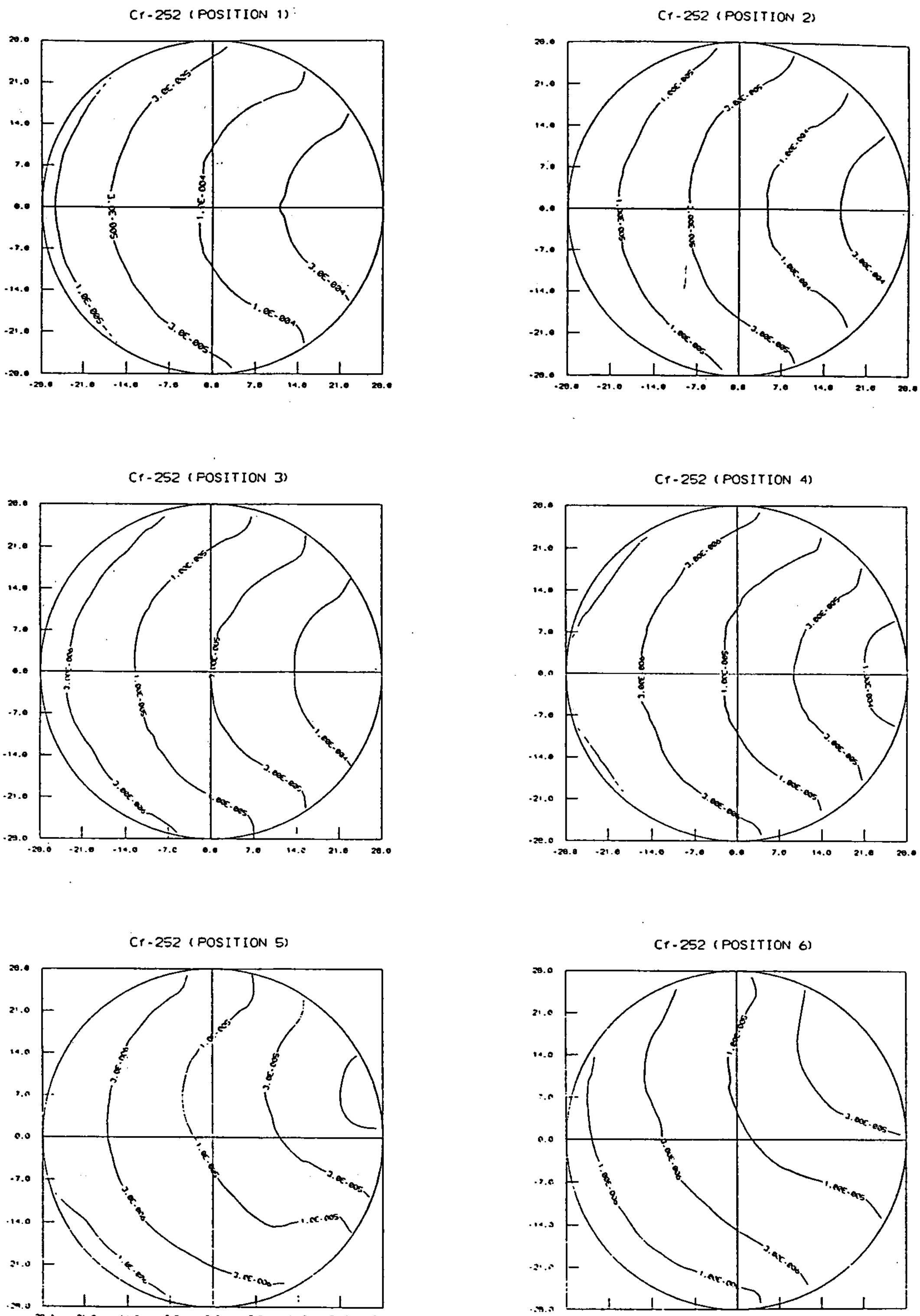

Figure 8 2D neutron-flux distributions of the thermal neutron flux (^{252}Cf source, central segment)

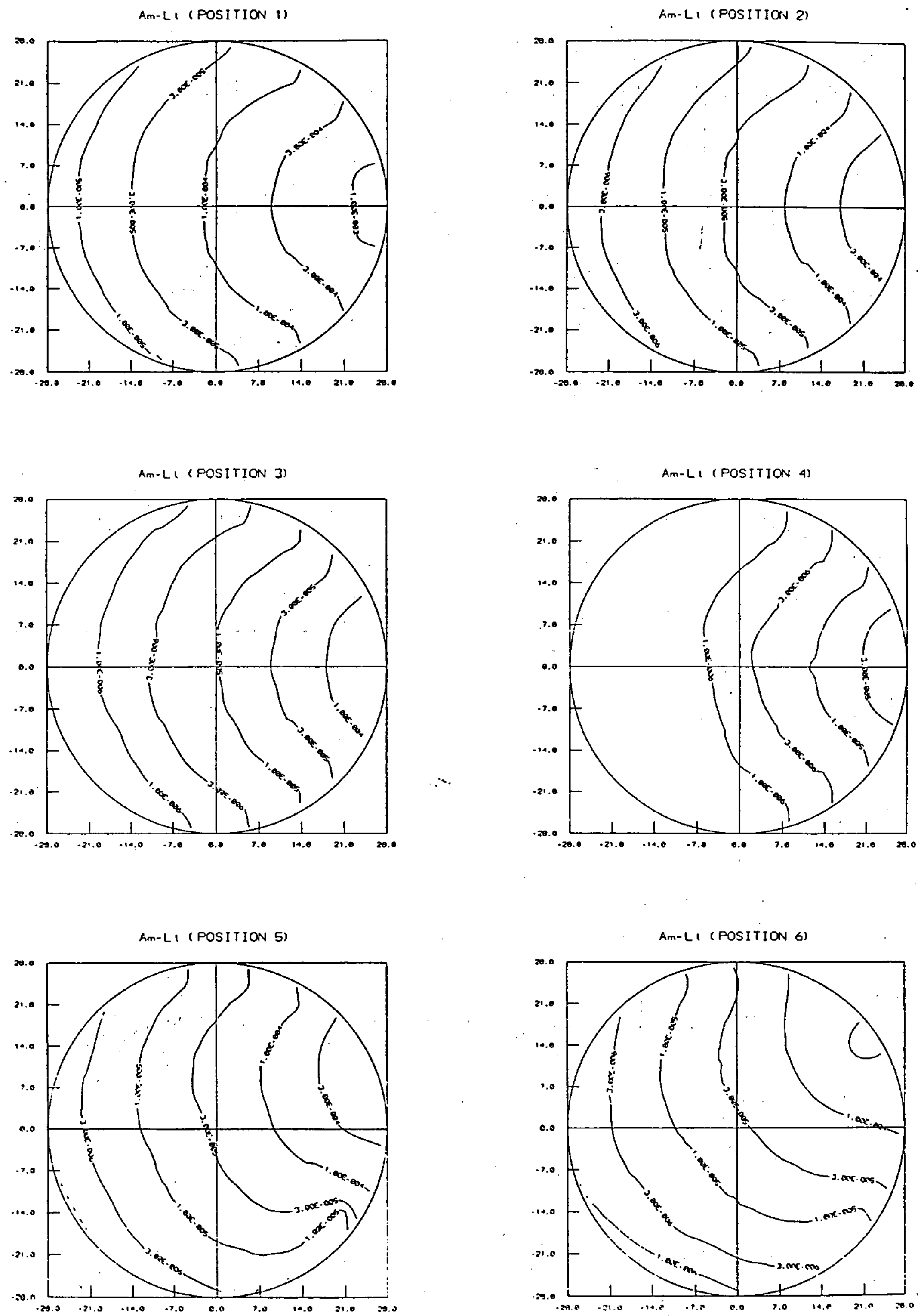

Figure 9 2D neutron-flux distribution of thermal neutron flux (Am-Li source, central segment).

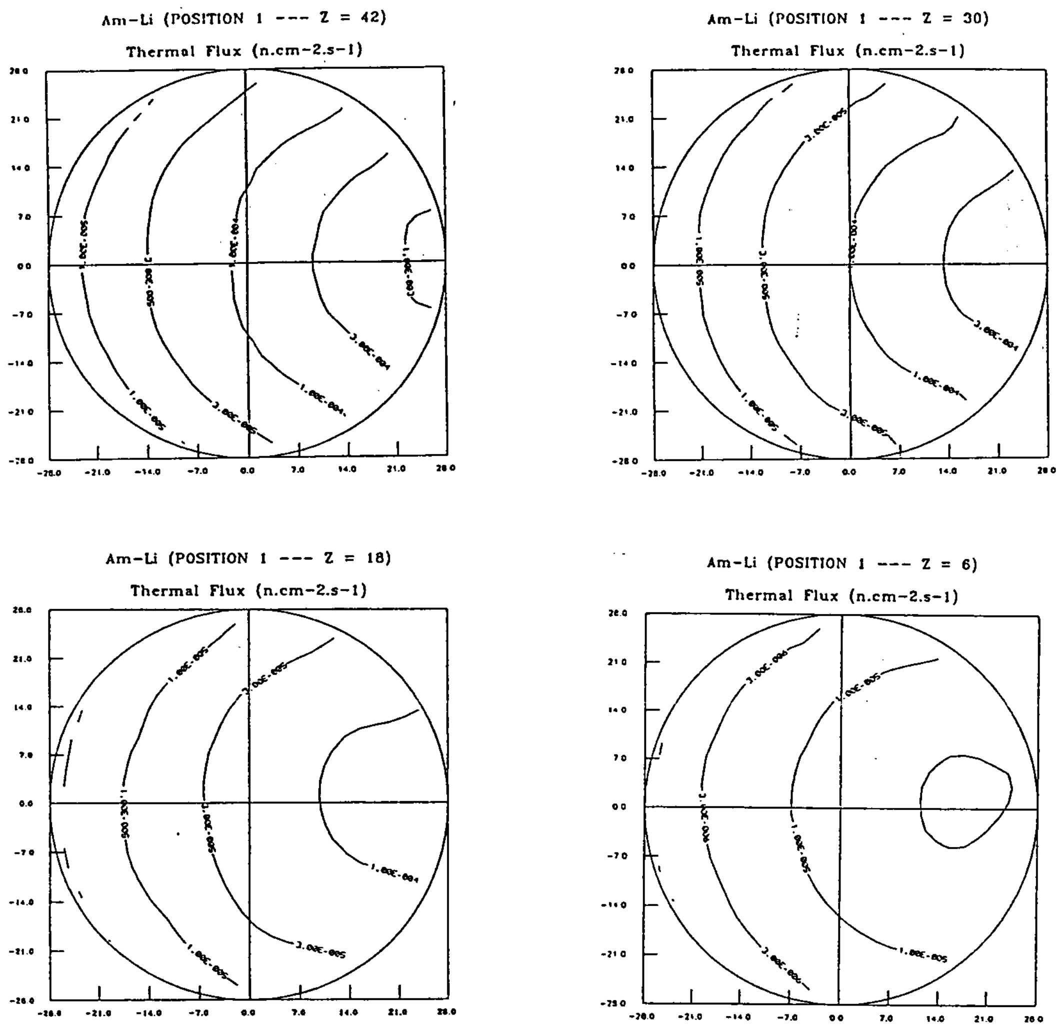

Figure 10 2D neutron-flux distributions of thermal neutrons (Am-Li source, position P1) in segments at Z=6 cm, Z=18 cm, Z=30 cm and Z=42 cm.

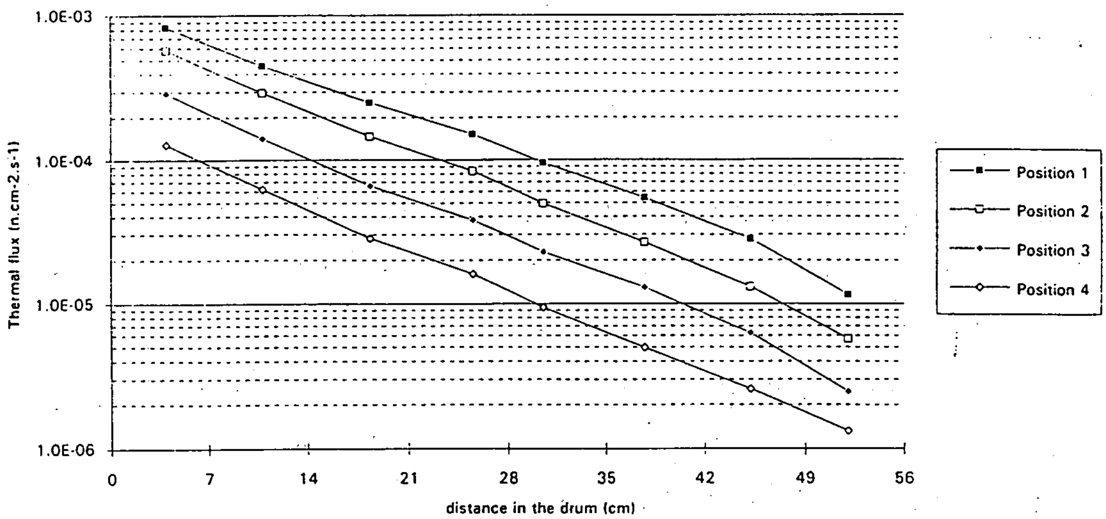

Figure 11 Variation of the thermal-neutron flux along the axis source-drum-centre for the C-252 source.

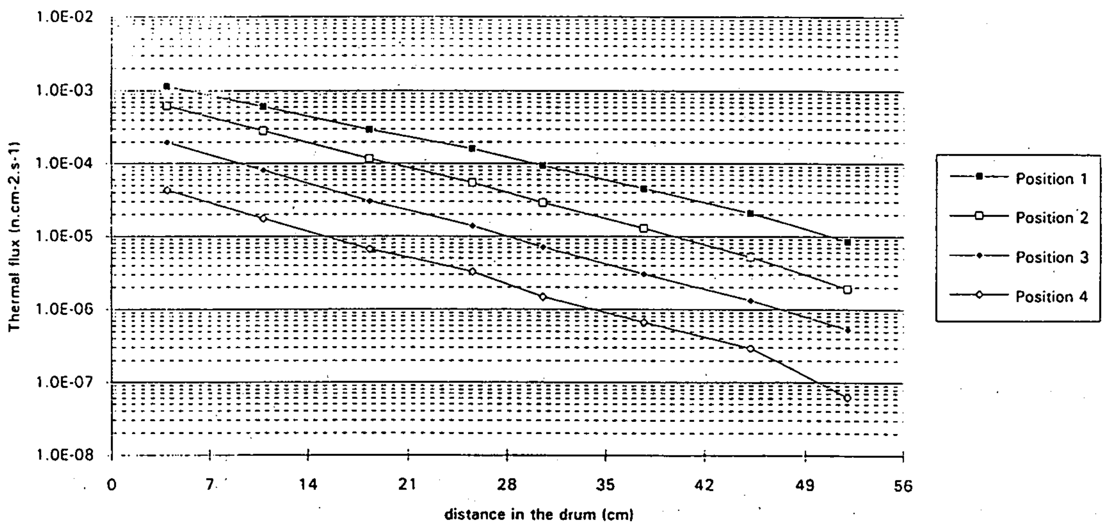

Figure 12 Variation of the thermal-neutron flux along the axis source-drum-centre (Am-Li source).

F Detector response for the active system in the DSB configuration

This series of Monte Carlo calculations evaluate the detector response of the DSB configuration of the active waste assay system as a function of the fissile material (^{235}U) uniformly distributed in the 220 l drum for three different neutron source spectra: Am-Li, Sb-Be and an accelerator spectrum corresponding to neutron energies between 30 and 100 keV. The accelerator spectrum was considered here because at the KFA experiments are planned with an accelerator as neutron source. Spectrum tayloring with the aid of moderators between source and waste drum was not considered here as it was proven in a previous paragraph that this will decrease the number of induced fissions in the waste package with a concrete matrix. The considered neutron sources were put at a distance of 10 cm from the drum wall. The ^{3}He detector (active length 50 cm) was positioned in the centre of the section of a 10 cm by 10 cm PE block with a length of 50 cm. The distance between detector and source was 10 cm and the space between source and detector was completely filled with PE. The results of the calculations with the MCNP-4.2 code are summarized in Figure 13, from which the following conclusions can be drawn:

- The detector response (this is the number of n-p reactions in the detector tube per source neutron) as a function of the mass of fissile material in the waste package can be described as a linear function of the ^{235}U mass with a slope different from 0;
- The background of source neutrons given by the respective intercepts of the curves with the y-axis clearly depends on the type of source spectrum. The Am-Li source — as could be expected— induces the highest background, while the Sb-Be source yields the lowest background.
- The three considered sources result in a comparable variation of the detector response proportional to the fissile mass.

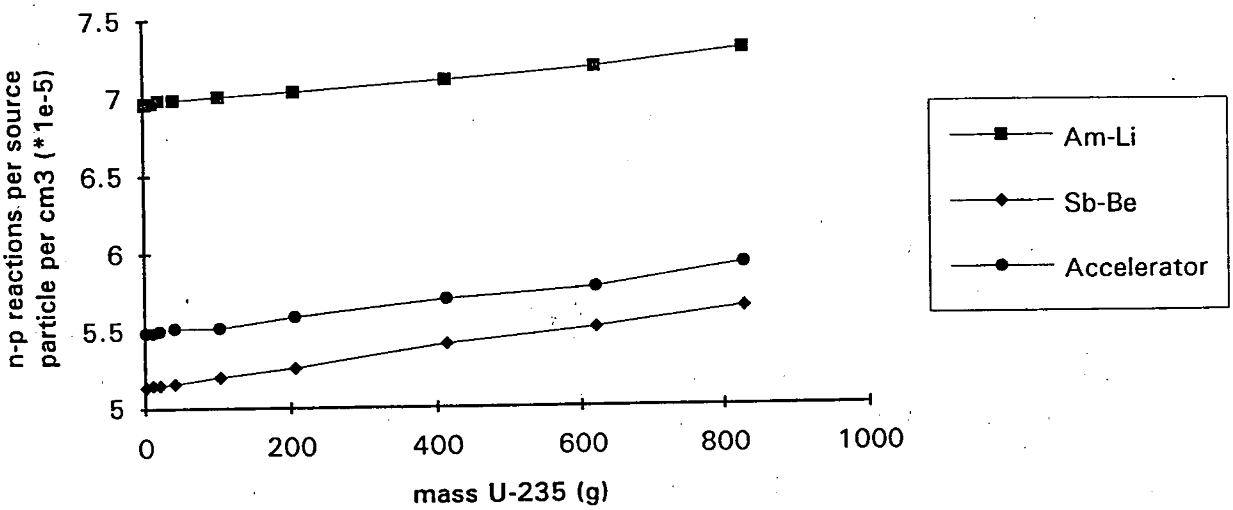

Figure 13 Detector response (n-p reaction in the detector per source neutron and per cm^3) for the active system (DSB-configuration, Am-Li source)

From the results of figure 13 the minimal detectable mass ^{235}U can be inferred by evaluating the source background (the principal source of noise) and the response (the signal) to the presence of fissile material (^{235}U). The minimum detectable mass is usually defined as that amount that corresponds to a signal three standard deviations above background. The minimum detectable mass clearly is a function of the source intensity (number of neutrons per second) and of the considered measuring time. We propose the following formula to evaluate the minimum detectable mass ^{235}U :

$$x = \frac{3\sqrt{b}}{a\sqrt{C}}$$

In which:

- b is the neutron background (the intercept on the y-axis of figure 13) ;
- a is the slope of one of the curves of figure 13;
- C is the product of measuring time (s), detector volume (cm^3) and source strength (n/s);
- x is the minimum detectable ^{235}U mass in g.

For the particular case of 900 s measuring time, a detector volume equal to 245.43 cm^3 (the detector volume considered in the calculations) and a neutron source intensity of 1E6 n/s, minimum detectable ^{235}U masses were calculated for the different source types; they are given in Table 6.

Source spectrum	Am-Li	Accelerator	Sb-Be
Min. Det. ^{235}U mass (g)	14.6	9.7	7.5

Table 6 Calculated minimum detectable ^{235}U masses (g) for the DSB configuration of the active system for 3 different neutron-energy spectra.

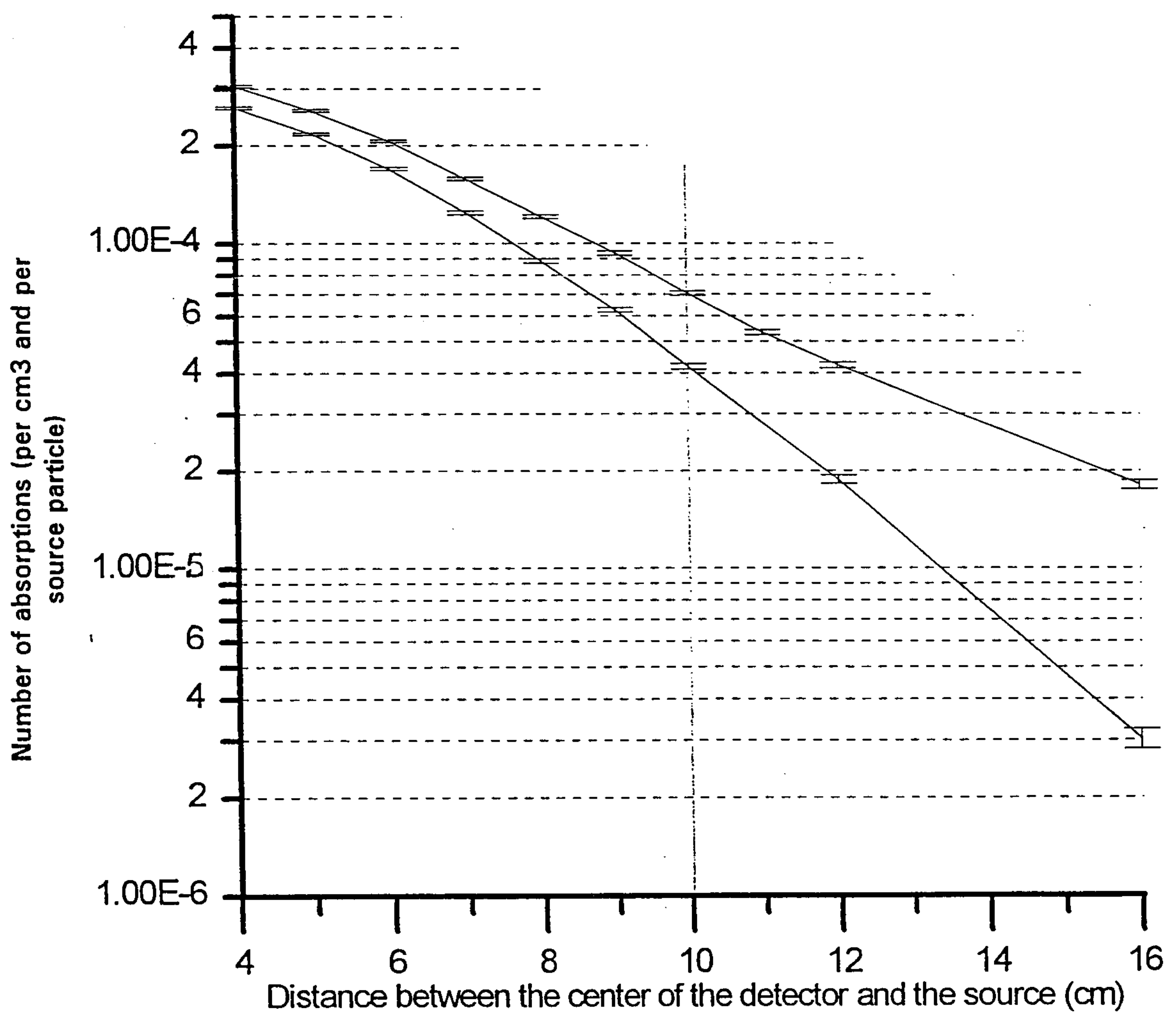

Figure 14 Detector response for the source background with (upper curve) and without (lower curve) reflections at the drum.

The detection limits clearly decrease when the source background decreases e.g. when the distance between source and detector increases. Figure 14 illustrates the variation of the background as a function of the distance between the source and the detector (the space between both is filled with PE). Figure 14 shows two curves; the lower curve corresponds to the source background when there is no drum in front of the detector-source assembly, the upper curve gives the detector response when a drum with concrete matrix but with no fissile material in it is placed in front of the assembly. An important contribution to the neutron background clearly arises from neutrons scattered by the waste package; this dominates for a thick PE layer between source and detector (see Figure 14). This perturbing contribution of neutrons cannot be decreased by improving the direct shielding of the source in the direction of the detector. As a consequence the net neutron background caused by the source only decreases slowly with increasing distance between source and detector tube. Increasing the distance from 10 cm (the detector-source spacing used to obtain the minimum detectable ^{235}U mass in the example above) to 16 cm will reduce the minimum detectable ^{235}U mass from 14.6 g to 7.5 g (Am-Li source at 1E6 n/s, 15 min. measuring time).

G Passive neutron counting

If the external neutron source is omitted in the active system, the detector block still can be used to detect neutrons from the spontaneous fission reactions and (α,n) reactions in the waste package. Normally passive neutron counting applied in the assay of nuclear waste uses several detector tubes to approximate a 4π detection geometry and to reach low detection limits. The single detector tube system used as a passive neutron counter is expected to have high detection limits and will limit its use to passive neutron measurements of waste packages with high plutonium content or high neutron output. It was expected that the performances of such a simple system would still be better than those of a conventional dosemeter. To estimate the detection limits of the one detector passive neutron system, a series of Monte Carlo simulations were initiated mainly to evaluate the detection efficiency of the system [Mandoki-93]. Considering the layout of the system, the following parameters were defined (see also Figure 15):

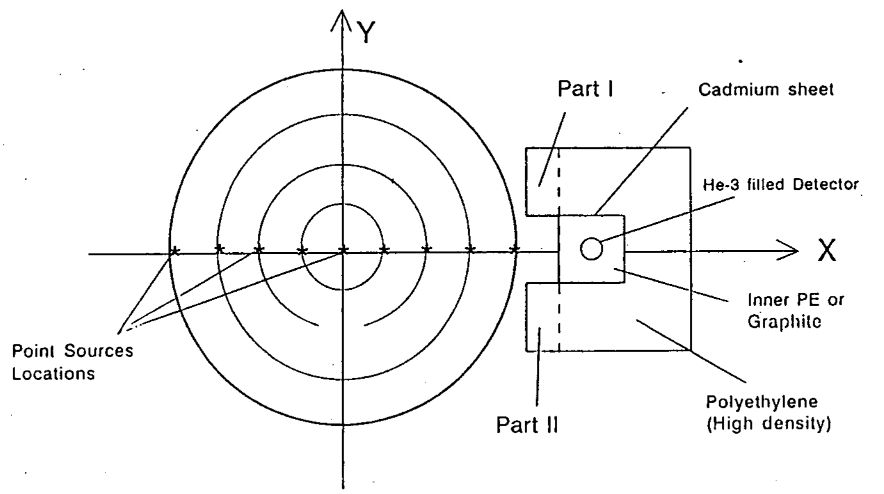

Figure 15 Detector-block configurations used in the calculations for the passive system.

- detector tube : He-3 at 10 bar pressure with an active length of 0.5 m and a diameter of 2.54 cm;
- moderator block: high density PE or graphite (with the option of a cadmium liner). The moderator block is surrounded by a 10 cm thick PE shielding except at the side in front of the waste package;
- waste package: 220 l drum with a concrete matrix (see [Mandoki-93]);
- fissile material distributions: point source or a homogeneous distribution;

In a first series of calculations the optimal dimensions of the moderator block were evaluated. The detection efficiency was calculated for different point-source positions in a concrete matrix and for different thickness of the PE moderator in front of the detector tube. The results of these calculations are shown in Figure 16. The point source was put at positions varying between x=28 cm and x=-28 cm on a line connecting the drum centre and detector centre. The positions x=28 and x=-28 cm respectively correspond to the drum wall close to and away from the detector. The value x= 0 corresponds to the drum centre. From Figure 16 it is clear that the optimal moderator thickness for the detector block depends on the position of the source. When the source is close to the detector an optimal thickness corresponds to 3 to 4 cm PE. When the source is moved towards the drum centre and beyond the centre x < 0, the detection seems to be optimal with a bare detector (no PE). The optimal thickness should be evaluated in a case by case study considering the thickness of concrete shielding in the waste drum and for volumetric source distribution.

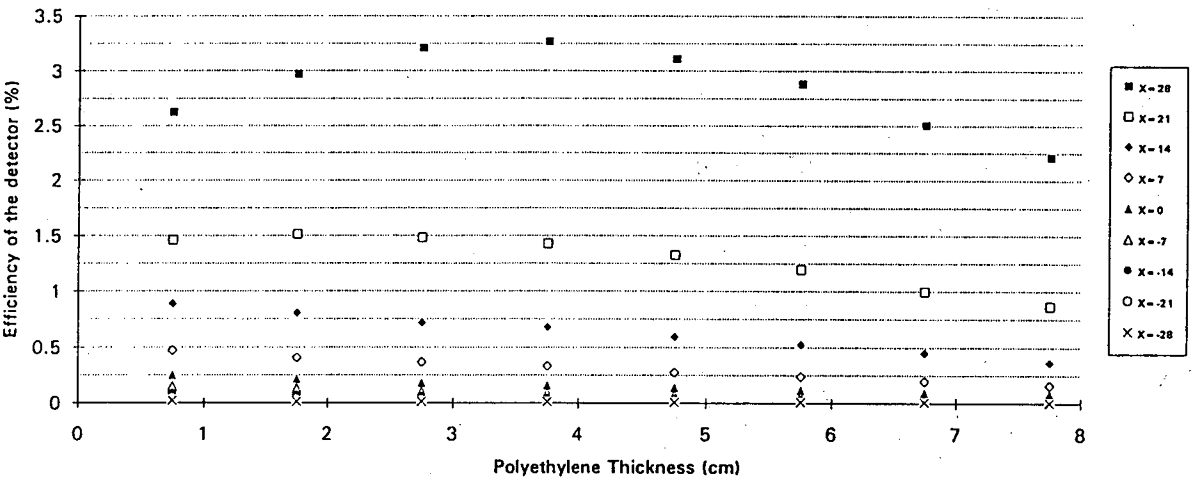

Figure 16 Detection efficiency as a function of the PE layer in front of the detector tube.

For the determination of the minimal detectable mass a fixed moderator thickness of 3.75 cm was considered and three different configurations of the detector block were evaluated (see Figure 15). The basic information to calculate minimum detectable masses is the detection efficiency of the system. The detection efficiencies for the configurations 1, 2 and 3 and for the specified characteristics of the detector block are given in Table 7.

	Configuration 1 detection efficiency (%)	**Configuration 2** detection efficiency (%)	**Configuration 3** detection efficiency (%)
detector fully surrounded by Cd	**0.12**	**0.13**	**0.15**
detector partially surrounded by Cd	**0.14**	**0.13**	**0.17**
Detector not surrounded by Cd	**0.15**	**0.16**	**0.21**
Detector surrounded by graphite	**0.29**	**0.28**	**0.40**

Table 7 Detection efficiencies for 3 configurations (see Figure 15) for passive neutron counting.

The highest detection efficiency of 0.40 % is obtained with configuration 3 and with a graphite moderator surrounding the detector tube. From the detection efficiencies minimum detectable plutonium masses can be inferred. Considering a neutron background of 0.03 counts/s (background measured at KFA Jülich for this system), a measuring time of 900s and a plutonium composition as given in Table 8.

Pu-isotope	Weight percent
^{238}Pu	1.21
^{239}Pu	61.84
^{240}Pu	23.79
^{241}Pu	8.91
^{242}Pu	4.25

Table 8 Plutonium composition used to estimate the minimum detectable plutonium mass.

If a detection efficiency of 0.15 % is used (this corresponds to the detection efficiency for one detector tube in a PE moderator), the minimum detectable plutonium mass equals 33 mg for total neutron counting. This estimation assumes no contribution from neutrons originating from (α,n) reactions. The exact contributions of (α,n) reactions to the total neutron output normally are only known in rare cases. These are also the only cases for which in total neutron counting the plutonium mass can uniquely be related to the neutron output. (α,n) reactions result in a higher neutron output for an equal quantity of Pu. This means that masses below 33 mg can be detected but the exact mass cannot be known unless the contributions of (α,n) neutrons are known.

H Conclusions

The principle of neutron energy-discrimination by means of energy-selective transport of neutrons through matter and the application of this technique in active waste assay systems was discussed using computer simulation of the neutron transport. Following the work programme and later discussions with KFA Jülich, mainly the Am-Li neutron source was investigated for its applicability in neutron interrogation waste assay systems. Two geometrical configurations for neutron interrogation waste assay systems were considered: the configuration referred to as the SBD-configuration which puts the waste package between source and neutron detector and the DSB-configuration in which source and detector are both at the same side of the waste package. Characteristics of both configurations were calculated by the use of the computer codes DTF-IV and the Monte Carlo code MCNP-4.2. Especially the influence of the neutron source was investigated. The Sb-Be source was already successfully used in a neutron interrogation system in the SBD configuration, but it needs severe gamma shielding and has a short half-life restricting its use in assay systems. Therefore the Am-Li source was considered as an interesting candidate source for neutron interrogation of waste packages.

The neutron-energy spectrum of the Am-Li source and other sources that might be considered emit more high-energy neutrons than the Sb-Be source. This undermines the first principles of energy-discrimination through selective neutron transport. For the Am-Li source in the SBD configuration, the calculations show that the signal to noise ratio of neutron counting can be optimized using spectrum tayloring in combination with efficient selective transport through polyethylene layers (selective transport in graphite has proven to be generally less efficient than in polyethylene). Up to now no detection limits were calculated for this configuration so that direct comparisons with the Sb-Be source or DSB configuration are not yet possible. The calculations of the spectrum tayloring of the Am-Li source and the selective neutron transport in the PE detector block have shown interesting and promising features with respect to neutron counting. Future calculations should evaluate detection limits for the Am-Li source in the SBD configuration.

The DSB configuration was thought to be more promising because this configuration has the neutron detector close to the well-interrogated part of the waste drum. This was the reason for paying special attention to this configuration. To function well the DSB-configuration needs an efficient neutron shielding in the direction of the detector. Calculations proved that a PE shielding combined with a cadmium-lined detector block provide the best shielding results. The calculated minimum detectable ^{235}U mass in a concrete waste form is 10 g for this configuration with an Am-Li interrogation source with an intensity of 1E6 n/s and a measuring time of 900 s. This rather high detection limit is due to an elevated neutron background from scattering of the source neutrons at the waste package. This background is dominant especially when the direct shielding between source and detector is good. With respect to this problem and the optimization of a neutron interrogation system with an Am-Li source it is worthwhile investigating a configuration in which this scattering is minimized by gradually moving the neutron detector from the DSB detector position to the SBD detector position.

According to the actions outlined in the work programme of the contract, we also investigated the passive neutron counting capabilities of the simple neutron counter in the total-neutron counting mode. The Monte-Carlo calculations show that a minimum detectable plutonium mass of the order of 30 mg can be achieved.

Comparisons between numerical results and experiments were not reported here mainly due to the fact that experiments at KFA were delayed caused by the non availability of the different neutron sources. These numerical results and corresponding conclusions however still may be verified experimentally by KFA on a later date. A direct comparison between computer simulations and experiments in the future also might be an important tool for the validation of real waste assay systems.

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