ACTIVE NEUTRON INTERROGATION DEVICES FOR GAMMA IRRADIATING SOLID ALPHA WASTES

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ABSTRACT

The paper presents experimental results concerning the measurement of the fissile mass content and the determination of alpha activity in solid waste having a strong gamma emission due to fission or activation products (800 l drums containing hulls,100 l drums).

In order to obtain accurate measurement,the passive neutron counting is combined with active neutron interrogation :252 Cf source with delayed neutron detection and differential die away technique using the SODERN GNTO2 neutron generator (prompt neutron detection between the 14 Mev neutron pulses.

INTRODUCTION

The accurate measurement of the fissile mass content and alpha activity of alpha radioactive wastes is necessary,particularly for the contribution to the fissile mass balance, for checking that the fissile amount is less than a maximal value (safety aspect) and to determine if the waste is in accordance with the surface storage limits.

Measurement methods using detection of radioactive emission are of special interest because they allow the determination of uranium or plutonium without altering the material.

For gamma irradiating waste, only the methods based on neutron detection apply.

The development of active interrogation techniques at the CEA/CADARACHE has been influenced by the following facts:

- in active interrogation, the determination of the fissile mass is very much less sensitive to an uncertainty on the isotopic composition than in passive neutron counting
- For Uranium contaminated wastes the active method is the only way to measure accurately the 235 U content.
- The low level of the surface storage criteria in FRANCE(0.1 CIα/T for acceptance,0.01CIα/T mean value on the storage site), and the great number of drums to be measured necessitate low detection limit systems (In the following,the detection limit is defined as being equal to twice the mass that gives a relative statistical error of 100 %,at a confidence level of 95 % (2σ)).

In order to have a better efficiency, he3 counters (working in pulse current mode) are used.

When the gamma dose rate at the level of the counters is greater than few rads/h a shielding (usually lead) is neccessary, but the presence of lead alter the performances of the method

In the paper we present experimental results obtained on experimental facilities at the CEA/CADARACHE in the NDA of solid gamma irradiating waste using the Californium shuffler, 14 Mev neutron generator,and also passive neutron emission measurement.

NEUTRON SOURCE METHOD USING DELAYED NEUTRON COUNTING

Physical Principle

The Californium source is alternatively introduced inside the cavity (about 10 s per cycle), to induce fissions, and removed in an isolated storage position (about 10 s per cycle).

During irradiation, fission fragments are produced and some of them decrease by emitting neutrons. In "thermal" interrogation, the cavity is composed of light elements in order to slow down neutrons from Cf and to maximize the fission rate.

When the source is in the storage position, the delayed neutrons are counted.

The delayed neutron signal may be considered as the sum of six decreasing exponentials, with a half life between 0.2 and 60 s, and mean energies between 25 and 500 kev.

The signal processing consists of searching for an amplitude term and a background term, by fitting a sum of exponentials to the signal, the amplitude term being proportional to the fissile mass content.

THE DANAIDE EXPERIMENT FOR 800 L DRUMS CONTAINING HULLS

The delayed neutron method, with 252Cf source has been recently applied to the measurement of 800 liter drums containing hulls (parts of fuel elements after cutting and dissolution of fuel assemblies in a reprocessing plant) in order to complete the passive neutron counting method for the determination of the residual fissile mass content and the alpha activity.

In passive mode, the neutron emission is mainly due to the spontaneous fissions of the 244 Cm.

The statistical error is low,but the main problem is that the specific neutron emission of the fuel is proportional to about the power 4,5 of the fuel burnup, and an imprecision on the burnup knowledge causes important errors on the determination of the fissile mass and the alpha activity.

Table I below shows the variation of the neutron emission and the alpha activity for various burnup.

TABLE I

Neutron Emission And Alpha Activity For PWR Fuel

Burn Up (MWD/T)	Neutron Emission (n/s/T)	Alpha Activity (Ci/T)
12000	$3.3\ 10^6$	$7.5\ 10^2$
20000	$3.1\ 10^7$	$2.0\ 10^3$
28000	$1.5\ 10^8$	$4.4\ 10^3$
33000	$3.2\ 10^8$	$6.7\ 10^3$

For a typical PWR fuel element the burnup is about 12000 mwd/t at the extremities and 33000 mwd/t for mean value.

Even if the burnup has been measured with a good precision at the entrance of the reprocessing plant,as it is not possible to certify the uniformity of the dissolution process, it is necessary to multiply the result by an important penalty factor to certify the result.

The interest of the active method is to directly measure the quantity of fissile material independently of burnup (for example low irradiated parts mixed with very well dissolved highly irradiated parts not "seen" by the passive counting because of low neutron emission for low burnups).

For the alpha activity, the combination of the passive counting(giving the neutron emission) and the active method (giving the fissile mass) is used to certify that the alpha activity is under a given value.

Mathematical modelling is apply in order to reduce the uncertainty on the location of the neutron emitters .

The experimental system is described in Fig. 1.

In this configuration, 10 cm of lead protect He3 counters from the high gamma dose rate ,in the case of a real drum.

The cavity is 2m high and 18 He3 counters (1 m long) are on the upper part, 18 on the lower part.

A modulated 252 Cf source (7.10^8 n/s) moved by a mobile shuffler unit was positioned on the top of the count-

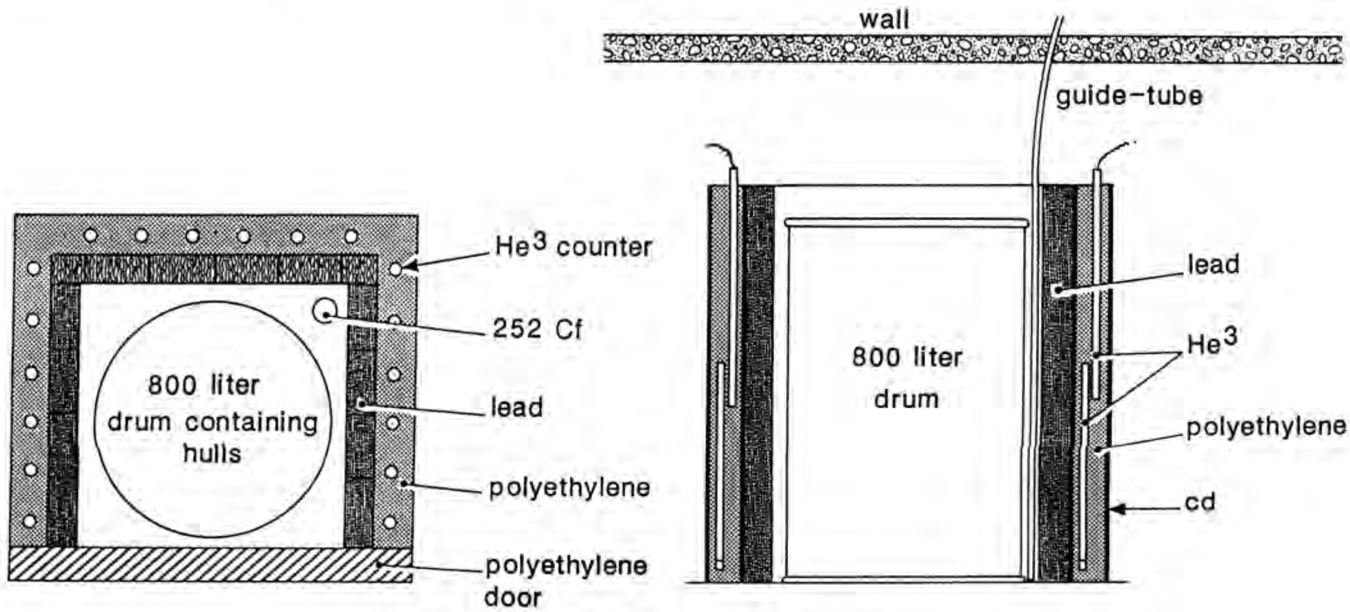

Fig. 1. Danaide Facility at CEN Cadarache.

ing cell. Measurements have been made by positioning fuel pellets at several axial locations.

On the horizontal plane, pellets were installed at 19 different locations to simulate a homogeneous repartition, the drum being filled with inactive hulls.

Figure 2 represents the variation of the utile signal, due to induced fissions,for five axial positions, with near homogeneous radial repartition.

We have also checked the ability of the signal processing to recognize the utile signal (decreasing exponential) with an important background (amplitude term at the origin of about 150c/s with a 2.10^4 c/s neutron background).

From these experimental results we obtain a detection limit, for a real drum, of about 6 g of fissile mass, for a 3 hours measuring time and a ^{252}Cfsource emitting 3 10^9 n/s. (and a neutron background of about 10^4 c/s).

According with the definition given in the introduction, the detection limit is proportional to about six times the square root of the background.

With 2 Californium sources (1.5 10^9n/s each) the effect of a non homogeneous radial repartition will be limited to about 20%.

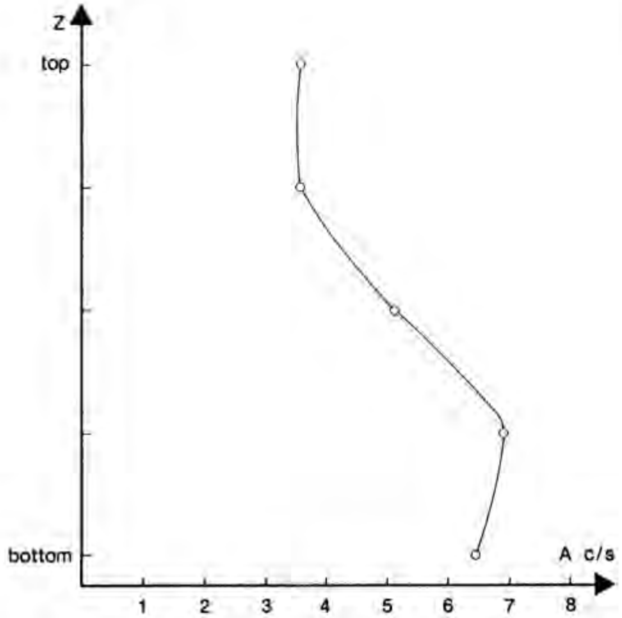

Fig. 2. Danaide Axial Response for Homogeneous Radial Repartition.

MEASUREMENTS SYSTEMS WITH 14 Mev NEUTRON GENERATOR

Physical Principal

In the differential die away technique, the 14 Mev neutrons produced in a short time (≈15 microseconds) by the neutron generator are slowed down inside the cavity and induced fissions.(after ≈0.5 ms the fast neutrons produced by the generator are in the low energy region).

The low energy ("thermal") neutrons flux decrease in time with a half life depending on the cavity and the matrix (near 1 ms in the PROMETHEE cell, in a empty cavity).

The he3 detectors are surrounded by a low energy neutron absorbing sheet, to detect essentially the fast neutrons (≈2 Mev) emitted when a fission event occurs.

The number of neutrons emitted in coincidence with the fission process (prompts neutrons) is several hundred times greater than the number of delayed neutrons and consequently this method has a lower detection limit than the detection of delayed neutrons with Cf source, for a same interrogating neutron source emission .

For gamma irradiating drums ,and for He3 detectors (1 m lengthg) working in pulse current detection, a protection (usually lead) is necessary to have a gamma dose rate, at the location of the detector lower than about 2 rads per hour.

Interaction of energetic neutrons with lead increase the neutron background (partially by the inelastic diffusion process) and deteriorate the detection limit.

The pulsed neutron generator method has several potentialities:

- the time decrease of the signals may be correlated to the matrix composition
- In passive mode, the combination of signals from detectors surrounded or not by absorbing sheet may be used to check if the matrix corresponds to those used for calibrations.
- For important masses,where self shielding may occur,the combination of delayed/prompt neutrons or thermal/epithermal interrogation may reduce this effect, especially for matrix with a small hydrogeneous content.

PROMETHEE 1 FOR 220 L DRUMS

The PROMETHEE assay is a modular system for experimenting pulsed neutron generator techniques.

The cell (Fig. 3) accomodating standard 220 l drums is set up with panels mounted on rails, allowing easy structure modifications.The drum rotates during measurements (17t/mn).

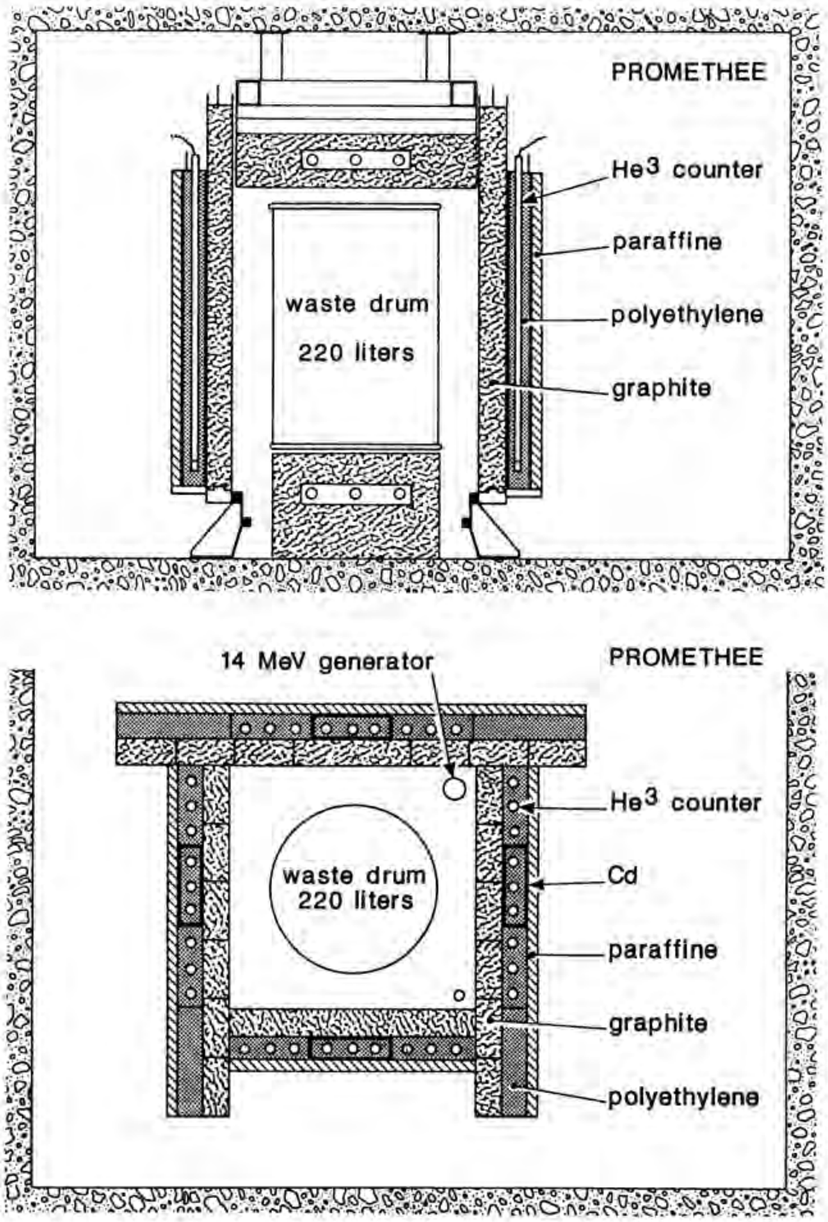

Fig. 3. Promethee Facility at CEN Cadarache.

The detection system includes twelve packages of three counters (1 m of utile length) on the lateral walls. Four of them are used for prompt neutron detection.

Three counters (45 cm of utile length) are positioned on the top and three others on the bottom of the cell.

A flux monitor is located at the corner of the cavity.

The neutron generator is the SODERN/GNTO2 ,working in pulsated mode (duration of pulse = 15 micro-seconds, time between pulses = 8 ms for the following results).

The amplifiers are outside the cavity, the pulses are counted on 8 MCS cards associated to a PC/AT computer.

The system could measure drums with a gamma dose rate up to 25 R/h without any modification on the signal.

This enables us, for example,the measurement of 220 l asphalt gamma irradiating drums. In this case the detection limit is 25 mg of 239 Pu, for a measuring time of 15 mn.

For a higher gamma dose rate a shielding is necessary in order to have a neutron counting rate independant from the gamma dose rate level.

Recent experiments have been made by surrounding a 100 l drum by 4 cm of lead, in order to determine the detection limit for an industrial system, having to measure drums with a gamma dose rate up to 500 R/h.

Experiments have shown that the absorption of low energy neutrons by the lead was approximatively compensated by the inelastic reactions (n,2n) and (n,3n) on lead.

The neutron flux is increased during the first milliseconds and decreases faster (absorption by lead); the "thermal" flux half time being reduced by 13 % (0.68 ms against 0.78 ms for an empty drum).

If in passive (global)counting the neutron background remains the same,in active mode the background is more important especially with inox steel matrix .

In the case of gamma irradiating drums the detection limit, for a 15 mn measuring time varies between 4.6 mg of 239 Pu (13 kg polyethylene matrix) to 17.4 mg (45 kg stainless steel matrix). Without lead the limits are respectively 2.8 and 5 mg.

PROMETHEE 2 FOR 800 L DRUMS CONTAINING HULLS

The PROMETHEE cell has been modified to experiment the DDT technique to the measurement of 800 l drums containing hulls, preliminary calculations having shown the interest of the method for UO2-PUO2 Mixed OXyde(MOX) fuels, having a neutron emission about 20 times greater than for Uranium OXyde fuels,the aim being to detect some grammes of residual fissile mass .

The size of the cavity (Fig. 4) is the same as for the DANAIDE experiment.

Six blocks of three counters (1 m length) are on three lateral walls, three of them on the upper part and three of them on the lower part.

The graphite walls are replaced by 10 cm of lead.

Measurements have been made with ^{235}U positioned at 19 different locations on the horizontal plane,the drum being filled with inactive hulls.

The figure 5 gives the net signal (c/s/g ^{235}U), for three axial planes (top, center and bottom).

The uncertainty (2 standard deviation)is about 3% for each value.

From experimental results we obtain a detection limit, for a real drum, of about 0.6g of fissile mass, for 3 hours measuring time, for a typical UOX fuel and 2.5 g of fissile mass for a MOX fuel (with a neutron emission 20 times greater than in the Uox fuel).

The maximum error, due to a non homogeneous repartition is about 50%.

CONCLUSIONS

The Active/passive systems that have been presented, for gamma irradiating wastes, met the criteria for fissile material and alpha activity measurement.

The neutron generator method that gives also information about matrix characteristics is well adapted.

Our development work on the solid gamma irradiating waste field is presently focused on the qualification of interpretation software for reducing the errors due to a non homogeneous repartition.

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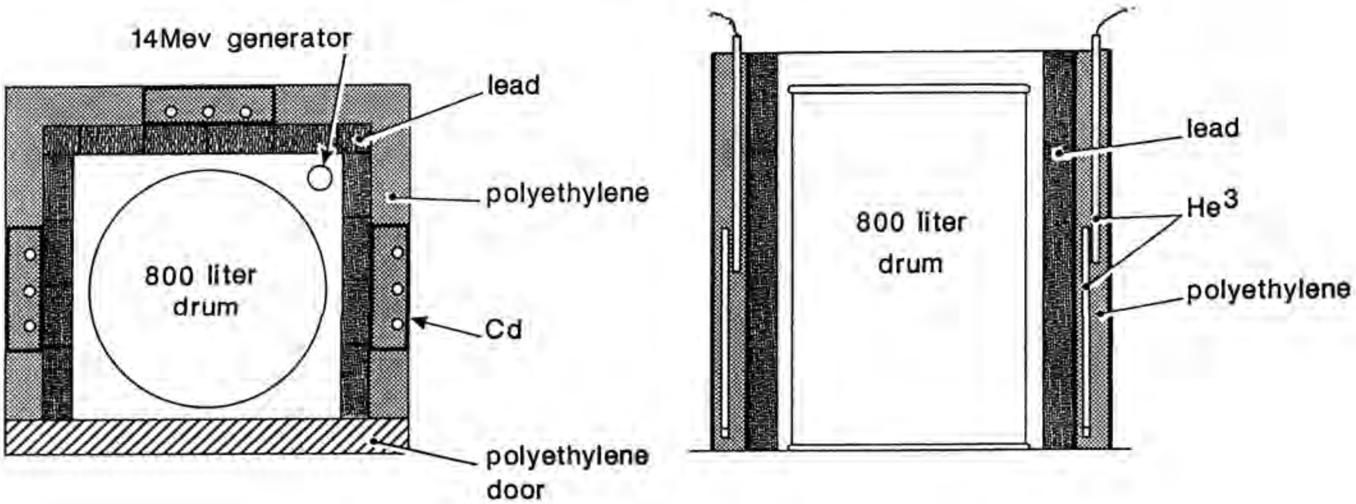

Fig. 4. Promethee Modification for 800 Liter Drums with Hulls.

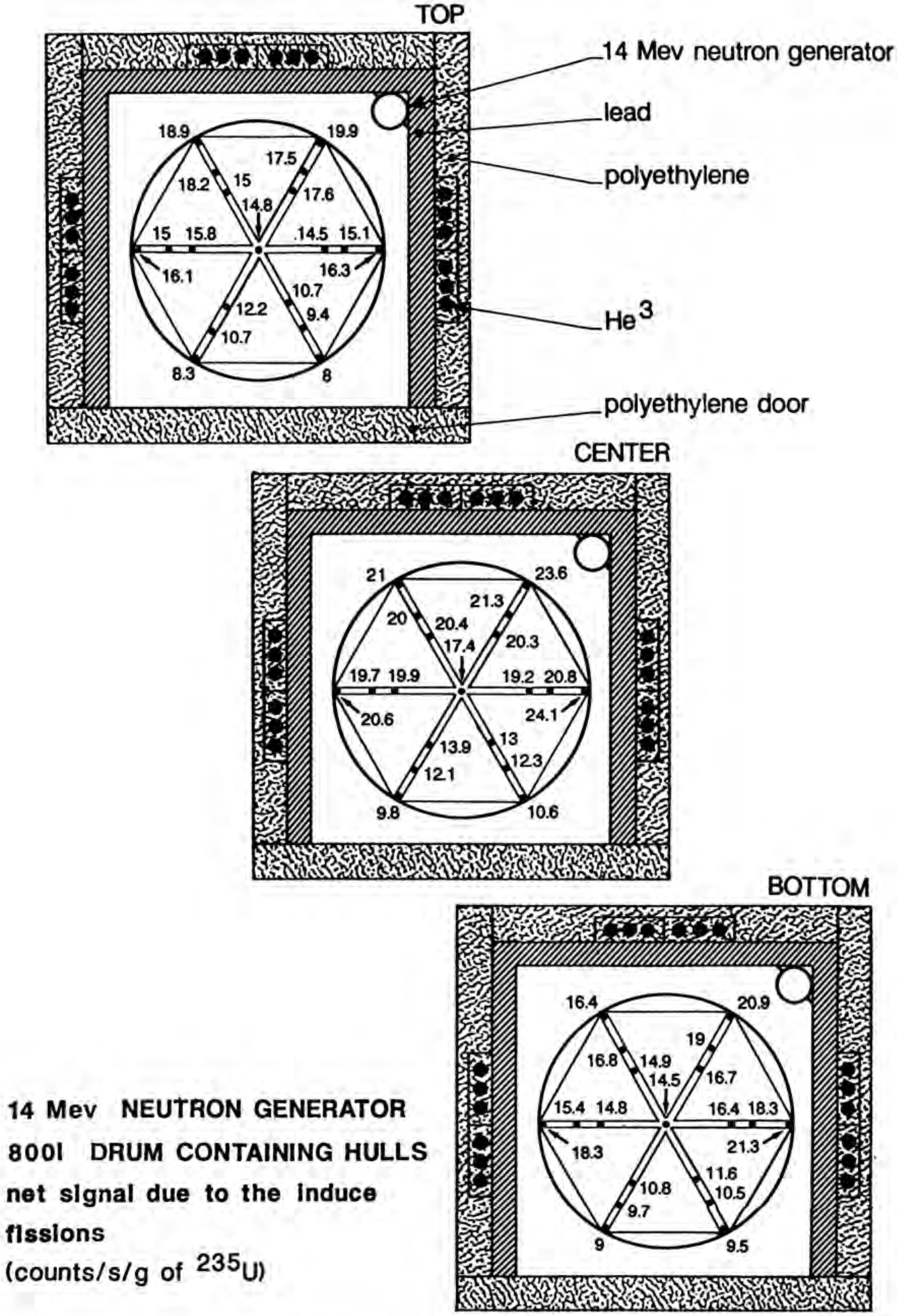

Fig. 5. 14 Mev Neutron Generator.

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