IEEE TRANSACTIONS ON NUCLEAR SCIENCE, VOL. 46, NO. 4, AUGUST 1999, pp. 877-879

Plutonium Detection With A New Fission Neutron Survey Meter

Alfred Klett
EG&G Berthold, P.O. Box 100163, D-75312 Bad Wildbad, Germany

Abstract

The search for illicit trafficking or hidden plutonium sources has become a pressing issue, especially since the breakdown of the former Soviet Union. Plutonium is extremely dangerous and hard to detect over large distances. The α-particles and X-rays which are emitted by plutonium isotopes can easily be shielded by the material itself or by surroundings. Besides a few γ's, the only penetrating radiation emitted by plutonium samples are neutrons from spontaneous fission.

Therefore a special neutron survey meter with unrivaled sensitivity for fission neutrons has been newly designed. The hand-held, commercially available instrument has an approximate weight of 4 kg and is battery driven. The neutron probe consists of a ^{3}He proportional counter tube, a moderator and integrated electronics. The sensitivity is sufficient to detect plutonium masses below 100 g at a distance of 1 m within a few seconds.

I. INTRODUCTION

There are several plutonium isotopes with long halflives, most of which are α-emitters. In addition to the α-particles, there is also emission of X-rays and of a few γ's [1], [2]. However, this is only weakly penetrating radiation and can not be used to detect plutonium over large distances or through shielding materials.

Plutonium isotopes with even mass numbers also decay by spontaneous fission and are thus neutron emitters. Fission neutrons are much more penetrating than α-particles or soft X-rays and they are well suited to give evidence of plutonium samples containing even numbered isotopes. For a plutonium survey with neutrons a detector with extremely high sensitivity for fission neutrons is required, because spontaneous fission rates are considerably lower than charged particles decay rates.

There are several types of counters available for neutron detection, but they are optimized to measure neutron dose rate. This is usually achieved by strong filtering with internal neutron absorbers. As a consequence these rem-counters are much too insensitive to detect sufficient amounts of plutonium within a short measuring time. Therefore, a special neutron survey meter with extremely high sensitivity for fission neutrons has recently been designed. The energy dependent response of this instrument was matched to the energy spectrum of fission neutrons [3].

II. SPONTANEOUS FISSION OF PLUTONIUM

The spontaneous fission yields Y_{SF} and the spontaneous fission neutron yields Y_{SFN} for plutonium isotopes of even mass number are summarized in Table 1 [1], [2]. The fission yields of the odd numbered isotopes are negligible. Q_N, which is the number of neutrons emitted per second and per unit mass, has been calculated and is given in the fourth column of Table 1.

Table 1
Spontaneous fission neutron emission of plutonium.

Nuclide	Y_{SF} [Bq^{-1}]	Y_{SFN} [Bq^{-1}]	Q_N [$g^{-1}\,s^{-1}$]
^{236}Pu	8.10×10^{-10}	1.81×10^{-10}	3560
^{238}Pu	1.84×10^{-9}	4.20×10^{-9}	2660
^{240}Pu	4.95×10^{-8}	1.09×10^{-7}	920
^{242}Pu	5.50×10^{-6}	1.23×10^{-5}	1790
^{244}Pu	1.25×10^{-3}	2.85×10^{-3}	1870

The total neutron emission of plutonium samples depends on the mass, the chemical form and on the isotopic composition. In plutonium mixtures there are usually large fractions of ^{239}Pu (50% to 95%) and smaller fractions of ^{240}Pu (5% to 30%), with a few percent or even less for the remainder. "Weapons plutonium" is relatively pure and has typically 94% ^{239}Pu with 6% ^{240}Pu, while "dirty plutonium" has larger amounts of even isotopes.

III. INSTRUMENT DESCRIPTION

To detect thermal neutrons a ^{3}He proportional counter tube is used. The counter tube has diameter 41 mm, active length 105 mm and overall length 222 mm. It is centered in a moderator made out of polyethylene with dimensions 310 mm $\times$ 180 mm $\times$ 80 mm. The moderator's and the counter tube's dimensions are optimized to achieve maximum sensitivity for fission neutrons. Preamplifier, discriminator and high voltage supply circuits are integrated in the neutron probe on top of the moderator. External dimensions of the neutron probe are 310 mm $\times$ 180 mm $\times$ 130 mm, and its total weight is only 3850 g.

Digital signals are transmitted to a separate battery driven µ-controller unit, which processes the detector signals. The reading of the instrument is shown on a dot matrix display. A sophisticated survey algorithm is used in order to detect even small changes in counting rate as quickly as possible.

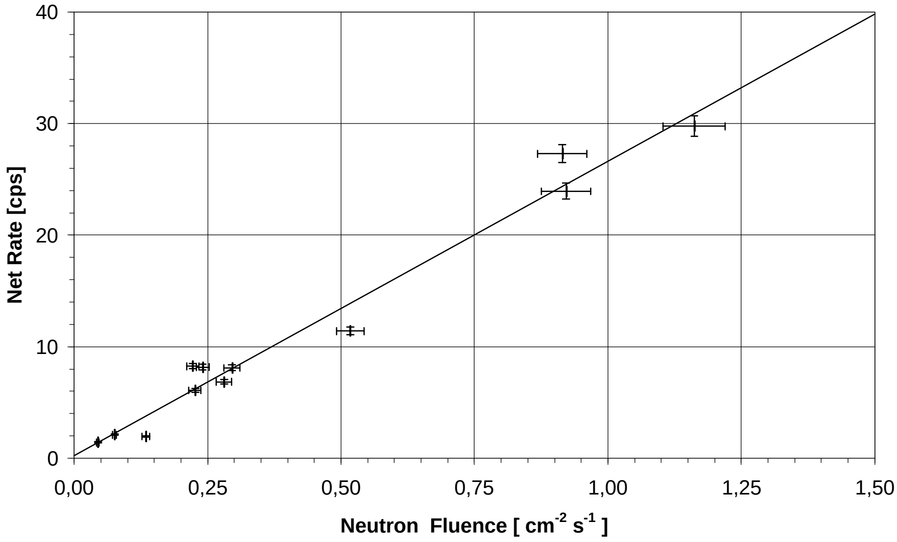

Figure 1: LB6414's fluence response to spontaneous fission neutrons emitted by plutonium samples.

IV. CALIBRATION

The instrument was calibrated with plutonium samples of different masses, isotopic compositon and chemical form in April 1998. These samples were provided by EURATOM in the Research Center Karlsruhe, Germany. Masses and isotopic compositions are listed in Table 2.

The gross rates measured at a distance of 1 m have been corrected for background and rescattered neutrons. The net rates are shown in Figure 1 as a function of neutron fluence, which was calculated with the parameters from Tables 1 and 2. The fluence response R_ϕ of the detector system is given by the slope of the straight line fitted to the datapoints. At 26.4 cm² the fluence response exceeds the Leake-counter's [4], which is a spherical standard type rem-counter, by nearly two orders of magnitude. It is also a factor of 25 better than the best currently available modern rem-counter [5].

Table 2
Plutonium samples with isotopic composition.

Mass [g]	^{238}Pu [%]	^{239}Pu [%]	^{240}Pu [%]	^{241}Pu [%]	^{242}Pu [%]
86	1.3	64.5	26.2	3.8	4.3
122	0.01	91.4	8.3	0.2	0.05
141	0.1	77.9	20.1	1.3	0.5
197	0.1	80.3	18.3	0.9	0.5
239	0.003	97.4	2.5	0.02	0.001
288	0.1	85.7	13.3	0.6	0.3
295	0.2	75.4	22.1	1.6	0.7
331	1.3	64.1	24.8	5.1	4.7
367	0.02	91.5	8.3	0.2	0.04
393	0.01	95.3	4.6	0.1	0.01
544	0.4	71.2	24.9	1.9	1.7
872	0.1	85.2	13.5	0.9	0.4

V. DETECTION LIMITS

As the neutron emission strongly depends on the isotopic composition of plutonium samples, two sample types have been defined. For further discussion in this paper, "weapons plutonium" is understood to be 6% ^{240}Pu with 94% ^{239}Pu, while "dirty plutonium" is meant to be 25% ^{240}Pu with 75% ^{239}Pu. It is much harder to detect "weapons plutonium" because its neutron emission rate is considerably lower than that of "dirty plutonium". With the values from Table 1 the neutron emission rates are 55.2 $g^{-1}\cdot s^{-1}$ for "weapons plutonium" and 230 $g^{-1}\cdot s^{-1}$ for "dirty plutonium".

For neutron detection an instrument's sensitivity for fission neutrons is specified in terms of ist fluence response R_ϕ. Fluence response is an effective cross sectional area of a hypothetical detector with 100% efficiency. With a fluence response of 26.4 cm² for fission neutrons the LB6414 offers an extremely high sensitivity. A neutron flux $\phi_n = 1\ s^{-1}\cdot\ cm^{-2}$ will generate a counting rate of 26.4 cps.

Besides the sensitivity of a detection system, the background counting rate also critically affects detection limits. In the presence of a natural neutron dose rate level of 8 nSv/h, the background counting rate of the Fission Neutron Survey Meter LB6414 is only 0.06 cps. As the average background level is so low, the observation of a few counts within seconds strongly indicates the presence of a neutron source.

Detection limits were calculated for both types of plutonium samples as a function of the distance between sample and detector. The detection limits were determined with a 95% confidence level with the measuring time as a parameter with values 2, 5 and 10 seconds. The calculation of detection limits follows the method described by L.A. Currie [6], and the results are shown in Figure 2 and Figure 3.

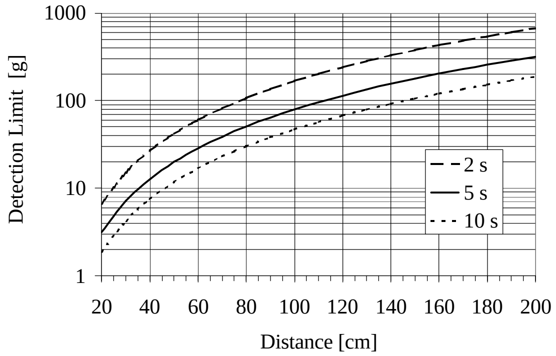

Figure 2: Detection limit for "weapons plutonium" as a function of distance for measuring time 2, 5 and 10 seconds.

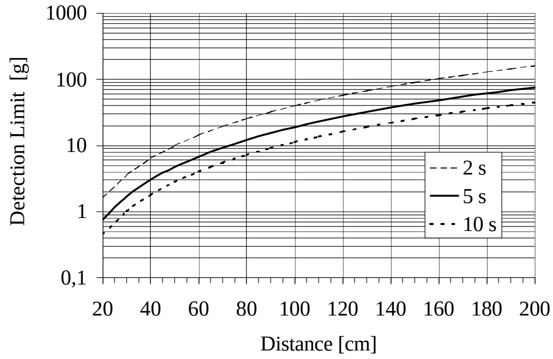

Figure 3: Detection limit for "dirty plutonium" as a function of distance for measuring time 2, 5 and 10 seconds.

VI. TECHNICAL SPECIFICATIONS

The commercially available Neutron Survey Meter LB6414 is a hand-held instrument, designed and manufactured in a ruggedized packaging. The system offers an acoustic alarm, an adjustable alarm threshold setting, memory for measured values, a scaler-timer function and an optical serial RS-232 port for ease of operation. The battery lifetime is > 150 h. Technical data are summarized below in Table 3.

Table 3
Technical data of neutron survey meter LB6414.

Neutron detector	^{3}He proportional counter -
Moderator	Polyethylene
Dimensions moderator	$310 \times 180 \times 80$ mm^3
External dimensions probe incl. electronics	$310 \times 180 \times 130$ mm^3
Weight	3850 g
Fluence response to fission neutrons	26.4 cm^2
Neutron energy range	Optimized to 10 – 1000 keV
Background rate	0.06 cps
Electronics	Integrated preamplifier and high voltage supply
Data acquisition	Datalogger LB1230
Display	32×64 dot matrix display
Data output	Optical RS232
Alarm	Acoustic, adjustable threshold
Power supply	Batteries or rechargeble batteries
Battery lifetime	> 150 h
Temperature range	-15°C to 50°C

VII. CONCLUSIONS

It is very hard to detect plutonium by its emission of charged particles or γ-radiation. In this paper it has been shown, however, that a well designed fission neutron survey meter provides extremely high sensitivity to plutonium. The instrument is not only capable of detecting hidden plutonium samples very quickly, but it can also detect plutonium contaminations in nuclear facilities or other neutron emitters like, for instance, ^{252}Cf or ^{244}Cm.

VIII. REFERENCES

[1] The International Commission on Radiological Protection, "Radionuclide Transformations Energy and Intensity of Emissions", *ICRP Publication 38, Annals of the ICRP*, Volume 11-13, 1983.

[2] C.M. Lederer and V.S. Shirley, Table of Isotopes, New York, John Wiley, 7th Edition, 1979.

[3] R.V. Griffith, J. Palfalvi, U. Madhvanath, "Compendium of Neutron Spectra and Detector Responses for Radiation Protection Purposes", Technical Reports Series No. 318, International Atomic Enery Agency, Vienna, 1990.

[4] J.W. Leake, "An Improved Spherical Dose Equivalent Neutron Detector", *Nuclear Instruments and Methods*, Vol. 63, pp. 329-332, 1968.

[5] A. Klett, B. Burgkhardt, "The New Remcounter LB6411: Measurement of Neutron Ambient Dose Equivalent H*(10) according to ICRP60 with High Sensitivity", *IEEE Transactions on Nuclear Science*, Vol. 44, No. 3, pp. 757-759, June 1997.

[6] L.A. Currie, "Limits for Qualiative Detection and Quantitative Determination", *Analytical Chemistry*, Vol. 40, pp. 586-593, March 1968.