Photoneutron Interrogation of Uranium Samples by a 4 MeV LINAC. A Feasibility Study

L. Lakosi, C. T. Nguyen, J. Bagi

Institute of Isotopes, Hungarian Academy of Sciences
P. O. Box 77, H-1525 Budapest, Hungary

E-mail contact of main author: lakosi@alpha0.iki.kfki.hu

Abstract. It is difficult to reveal smuggled U-bearing material by passive gamma-detection, because the weak radiation can easily be shielded. Neutrons, as penetrate shielding, represent a detection potential, by inducing fission in the nuclear material. A 4 MeV linear accelerator was used as a pulsed neutron source for active interrogation of U-bearing material. Produced in heavy water by bremsstrahlung, neutrons subsequently induced fissions in UO_2 samples. Delayed fission neutrons were detected in a neutron collar built up by four ^{3}He counters in a polyamide container. The counters were gated to be detached from high voltage during the electron pulse. Cyclic irradiation-measurement periods were subsequently used with a 25 Hz pulse repetition rate, as optimum setting, i. e. 500 cycles lasted for 20 s. The time analyser start-up was externally triggered and synchronised by the electron beam pulse. The response of the system was studied as a function of the intensity of the electron current, the amount of heavy water, U enrichment, and total U content. Sensitivity limit was achieved as 0.5 g ^{235}U and/or 30 g ^{238}U in a 20 s measurement time (500 cycles) with the amount of heavy water of 100 g and a mean electron current of 2 μA. Because of the long die-away time of neutrons, the duration of the interrogating neutron pulse is about 20 ms, so that a half of the time interval of 40 ms between pulses available for counting delayed neutrons can only be exploited. The response cannot be, however, enhanced by reducing frequency in order to lengthen counting time between pulses, because the saturation level of counting rate would decrease correspondingly. Nor the current is worth increasing, since the pulse tail would accordingly be lengthened.

1. Introduction

It is difficult to reveal smuggled U-bearing material by passive gamma-detection, because the weak radiation can easily be shielded. Neutrons, as penetrate shielding, represent a detection potential, by inducing fission in the nuclear material. Neutron generators using D(d,n)^{3}He, T(d,n)^{4}He [1-3], or ^{9}Be(d,n)^{10}B reactions [4, 5] are in common use for active neutron interrogation purposes.

Our aim was to establish, whether a low energy linear accelerator (linac) can be used as a pulsed neutron source suitable for active interrogation of nuclear material at border checkpoints. The advantage of accelerator use is that it can be switched off. Neutron production proceeds through conversion of the electron energy into bremsstrahlung and subsequent neutron production by photonuclear reactions. Use of heavy metals (Ta, W) is common as targets, whereas electron energies as high as 7-20 MeV are needed for photoneutron production [6], but 50-150 MeV beams are frequently used for achieving higher assay sensitivity [7].

Direct photofission is a special type of photonuclear reactions, where the nuclear material itself serves as target of a (γ,f) reaction induced by the photon burst from an electron accelerator. Such γ-ray interrogation can be performed by a 10-15 MeV linac [8, 9]. When fission occurs, both prompt and delayed fission neutrons are produced, and are multiplied by the fissile material. Upon detecting fission neutrons, distinction from interrogating neutrons can be made by delayed neutron counting. Typically only about 1 % of the neutrons emitted from nuclear fission are delayed neutrons, all others are prompt. By detecting the former, prompt neutrons are lost, but in the absence of the huge

interrogating neutron background, the sensitivity may still be high enough for performing the assay, in spite of losing 99 % of the fission yield.

2. Method

In our institute we have a linac of 4 MeV electron beam. For production of neutrons with such low energy electrons, attention is immediately focused on Be and D, owing to their low (γ,n) reaction thresholds of 1.67 and 2.22 MeV, respectively. While these thresholds are quite low compared to heavy elements, the cross sections for the (γ, n) process are also smaller by about two orders of magnitude than the giant resonance cross sections for heavy nuclei. It is important to recognise, however, that the neutron yield may be determined more by the atomic processes governing attenuation of γ-rays than by the (γ, n) cross section itself. Thus the mean free-path in a heavy element for γ-rays is limited by the atomic cross section.

For the lighter targets of Be or D, both the atomic and the nuclear cross sections are much smaller. However, the ratio of atomic to nuclear cross sections is not too different from heavy targets. Therefore, neutron yields as high as those from heavy metals are in principle possible if the target thickness is increased to one atomic mean-free-path. While the target thickness is much greater for the light element compared with the heavy element, the yield might be comparable depending on the details of the dependence of the (γ, n) cross section with energy, etc. In those experiments, where target size is not an important factor, the lighter targets of Be and D might compete [10]. Actually, neutron yields from these light elements can be roughly an order of magnitude higher than those from heavy W or Pb even at 10 MeV [6], while capital and running costs are much lower of course at the same time.

Our choice for the neutron converter was heavy water, because it does not imply any environmental risk. Neutron production is due to (e,γ) and (γ,n) double conversion. The neutron energy available from the D(γ,n)H reaction is 1.8 MeV at 4 MeV electron energy, but the mean neutron energy is around 0.9 MeV. Less work has addressed the assay of nuclear material by lower energy linacs so far. By 70 cm^3 heavy water as neutron converter a 10 MeV linac provided ~10^9 n/s/μA neutron yield [11]. A thermal neutron beam of 1.23×10^8 $n/cm^2/s/mA$ intensity was achieved by a 5 MeV linac at 50 cm distance from a BeD_2 target [12].

The relative yields of the six main groups of delayed neutrons from U-235 fission induced by fast neutrons are shown in Table I [13]. The relative intensities are also given, obtained upon multiplying the former values by the respective decay constants. Uncertainties are in parentheses. It is seen that the contribution of the three first groups predominates, i.e. the number of delayed neutrons practically goes into saturation in the first ten s of irradiation.

TABLE I: RELATIVE CONTRIBUTION OF THE DELAYED NEUTRON GROUPS AT SATURATION

Group	$T_{1/2}$ (s)	Relative yield (%)	Rel. intensity ($10^{-3}s^{-1}$)
1	0.179 (0.017)	2.6 (0.3)	100,7(15.1)
2	0.496 (0.029)	12.8 (0.8)	178.8(15.3)
3	2.23 (0.06)	40.7 (0.7)	126.5(4.0)
4	6.0 (0.17)	18.8 (1.6)	21.7(2.0)
5	21.84 (0.54)	21.3 (0.5)	6.9(0.24)
6	54.51 (0.94)	3.8 (0.3)	0.49(0.04)

3. Experimental Setup

Bremsstrahlung was generated on a 20 by 30 mm size Pt converter positioned at 3 cm distance from the exit window of the linac type Tesla LPR-4 at the Institute of Isotopes. The diameter of the electron beam was about 2 cm at converter distance. The energy distribution of the electrons is about 0.67 MeV FWHM at 4 MeV. The 0.9 mm thick converter provided complete stopping of the incident electrons. Electron pulses are produced in one of the two basic modes of operation; either single pulses are fired by an external trigger, or the pulses are produced continuously with a repetition rate of 50, 25, 12.5 or 6.25 Hz. The normal pulse duration is 2.6 μs. The peak intensity is 200 mA, while the mean current intensity is 26 μA at maximum. Energy stability is about 4 %. The total bremsstrahlung output with energy above 1 MeV was calculated to be about $2.4(0.3)x10^{13}$ and $7.3(1.1)x10^{12}$ photon/s above 2.22 MeV at full intensity in all directions [14]. The cup containing ~100 g heavy water was placed at the top of a neutron coincidence collar. The detector-moderator configuration consists of two concentric polyamide cylinders of an outer size of Ø200x420 mm. The inner cylinder of Ø55 mm forms a measurement cavity of wall thickness 10 mm for the material to be assayed. In between the two cylinders four proportional counters filled with ^{3}He gas to a pressure of $4x10^5$ Pa (4 atm) served as neutron detectors [15].

The schematic block diagram of the signal processing electronics is shown in Fig. 1. A 512 channel analyser was used in multiscaler mode of operation as a time analyser. Triggering the analyser was synchronised with the linac control command pulse. The gate circuit turned off the high voltage during the electron pulse. A single fast preamplifier of a time constant reduced to 2-4 μs was built for the four proportional tubes. The channel width of the time analyser was 100 μs.

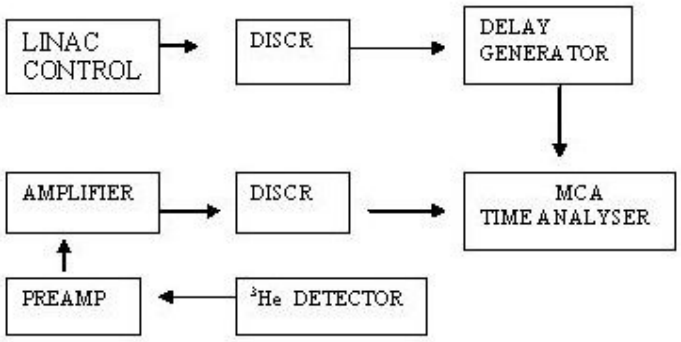

FIG.1. Schematic block diagram of signal processing

4. Measurements and Results

The delayed neutron signal from a 427 g UO_2 sample of 2.7 % enrichment was measured as a function of the pulse repetition rate, using one single counter tube only, at 4.4 μA mean electron current. The experiments were performed at 50, 25, 12,5, and 6.25 Hz. The neutron pulse tail was about ten ms long. Counting was started when primary + prompt fission neutrons already vanished, i. e. the effective counting times were 10, 30, 70, and 150 ms/pulse. The results for 200 irradiation-measurement cycles are summarised in Table II. It is seen that the number of delayed neutrons starts to grow at first, then decreases, as expected. The latter is due to the decrease of the saturation level as the pulse rate gets slower. A frequency of 25 Hz was established as an optimum. This frequency was used further on.

Assay of low enriched U samples was performed with 500 irradiation-measurements cycles, using four counting tubes, 103 g heavy water and a 2.2 μA mean electron current. The time spectrum of the detected neutron pulse is shown in Fig. 2. The initial 20 ms following the linac pulse is dominated by interrogating photoneutrons and prompt fission neutrons (in the presence of fissile material). In panel *a,* the time evolution of the neutron pulse is seen in the presence of heavy water, but with no sample in the cavity. In panel *b* the same is shown but with a UO_2 sample of 427 g, enrichment of 2.7 %. A counting period from 25 ms after the pulse up to the start of the subsequent pulse was established. In this period the count rate is practically constant, corresponding to the slowly decreasing count rate of the delayed neutrons at a saturation level, the latter governed by the pulse repetition rate. Net counts of delayed fission neutrons being equal to the difference of the two curves can be seen in panel *c*. The time interval suitable for measurement falls between T_1=25 and T_2=38 ms in the time spectrum, after vanishing the prompt neutron intensity. It is seen that prompt fission neutrons are measurable as well,

TABLE II: DELAYED NEUTRON COUNTS ACQUIRED IN 200 CYCLES

Frequency (Hz)	50	25	12.5	6.25
Total effective time for measurement (s)	2	6	14	30
Number of counts	50 (5)	90 (10)	85 (10)	60 (10)
Intensity (cps)	25 (2.5)	15 (1.5)	6 (1)	2 (0.5)

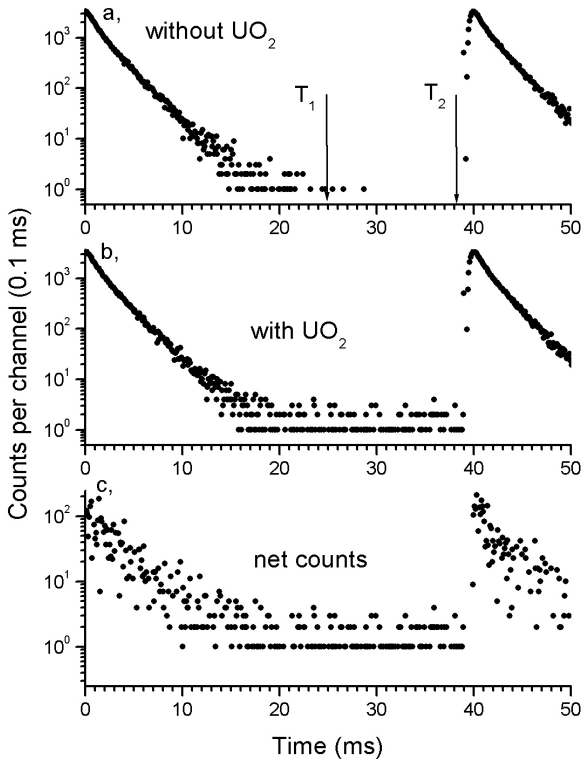

FIG. 2. Time spectrum of the detected neutron pulse

but they are an order of magnitude less than the primary interrogating (background) neutrons from the heavy water. The relative uncertainty (of a little difference of two big numbers) gets in this way large, until the lower boundary T_1 of the counting interval is reached. The counting interval from T_1 to T_2 is indicated for measuring delayed neutrons in Fig. 2, where there are no background (interrogating and prompt fission) neutrons any longer. The time T_1 is too long, it takes up more than one half of the time span between two pulses. The common feature of pulsed neutron systems is that the detected pulse lasts not only for the few μs duration of the inducing process, but even afterwards for thousands of μs [2, 8, 9]. The possible current intensity 26 μA cannot be fully exploited, because the duration of the long tail of a pulse depends on the amplitude, and by increasing the current, the duration would be even longer.

The response of the system was studied as a function of the intensity of the electron current, the amount of heavy water, U enrichment, and total U content. In Figs 3 and 4 the response to the electron current at 100 g heavy water and to the amount of heavy water at 4.4 μA mean electron current are indicated, respectively, using one single counter tube only.

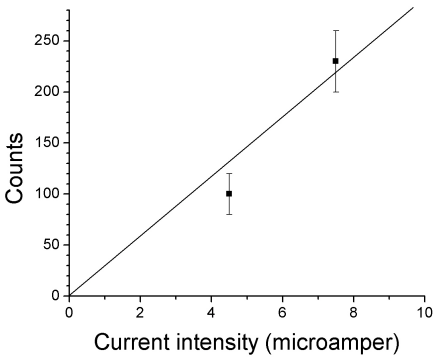

FIG. 3. Response to the electron current

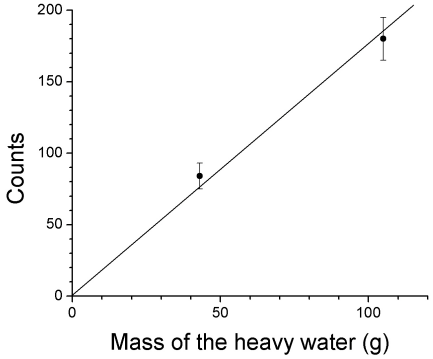

FIG. 4.Response to the amount of heavy water

Assay results of samples of four different enrichments are seen in Fig. 5. The lowest ^{235}U abundance is 0.28 % (depleted), the highest is 2.7 %. The masses of the samples were around 400 g. The responses are normalised to 400 g. It can be observed that ^{238}U also undergoes fission, actually its contribution already predominates even for samples of natural isotopic composition. This means that thermalisation is rather imperfect. The contribution of neutrons from ^{238}U fission can be read out from the curve. On the basis of these results, a sensitivity limit can be achieved as 0.5 g ^{235}U and/or 30 g ^{238}U in a 20 s measurement time (500 cycles).

In Fig. 6 the response is plotted as a function of the net ^{235}U mass. Result of a control experiment with a 90 % enriched U sample of 1.5 g total mass (^{235}U content ~ 1.126 g) is also indicated in this figure. It is seen that this test point fits well into the response curve.

The assay response of a series of samples of two ^{235}U abundances 2.7 and 0.28 % to the total U mass is shown in Fig. 7.

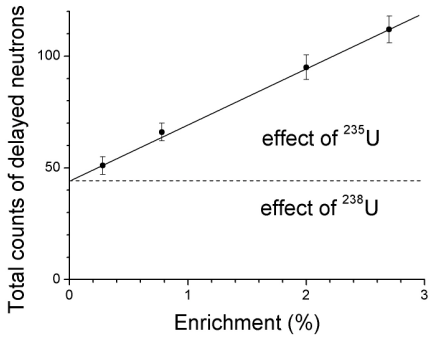

FIG. 5. Response to the enrichment

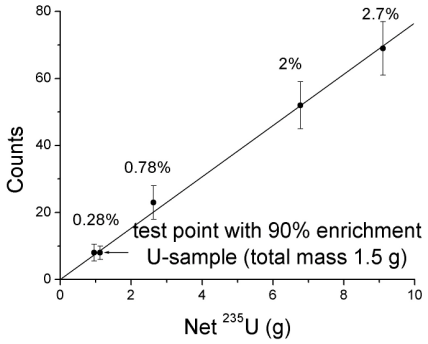

FIG. 6.Response to the ^{235}U mass

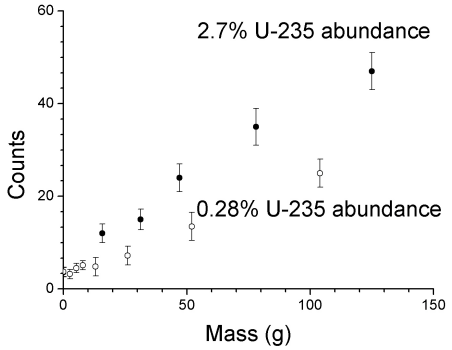

FIG. 7. Response to the total U mass

The optimum moderator thickness between the heavy water and the sample was established to be around 5 cm. By varying it, substantial sensitivity enhancement cannot be achieved.

5. Conclusions

During the accelerator pulse of the order of μs length, the detection system is paralysed for thousands of μs, and a long-tailed time spectrum is produced. Therefore a gate circuit was built which, triggered by the electron pulse, turned off high voltage. However, this did not affect the time constant of the long decay curve. Nevertheless, the gate circuit was used further on, in order to save the ^{3}He tubes.

The long tail cannot be affected electronically, so it is due to the long die-away time of interrogating neutrons. While in the neutron collar alone it is 70 μs for radioisotopic neutron sources [15], in this pulsed operation it is ~ 3 ms. Wrapping the container and the heavy water in Cd may help, – and this is planned to be tried - even though the response will reduce.

Owing to these circumstances, the performance of the accelerator cannot be fully utilized, i. e. there is no use of increasing the electron current, because not only the detected pulse amplitude grows up, but decay time of it also lengthens, reducing the interval available for counting. By extending the time between pulses, e. g. by halving frequency, saturation level and so the amplitude of delayed neutrons decreases proportionally, thus the number of counts – accumulated in a time twice as long - remains essentially unchanged.

In spite of the above difficulties, a sensitivity limit as 0.5 g ^{235}U and/or 30 g ^{238}U can be achieved in a 20 s measurement time (500 cycles) with the amount of heavy water of 100 g and a mean electron current of 2.2 μA. It means that this method is promising in respect of designing an active portal monitor for revealing unauthorised transportation of nuclear material.

Acknowledgement

This work was supported by the Hungarian Atomic Energy Authority under contracts No. OAH-ABA-76/00, OAH-ABA-ÁNI-05/01, and -05/02.

References

[1] ROSENSTOCK, W., et al., Measurement of delayed neutrons after induced fission of special nuclear material, Proc. IAEA Symp. Internat. Safeguards, IAEA, Vienna (1997) IAEA-SM-351/3P (CD-ROM).
[2] RAOUX, A.-C., et al., Transuranic waste assay by neutron interrogation and online prompt and delayed neutron measurement, Nucl. Instr. Meth. B 207 (2003) 186-194.
[3] BOGOLUBOV, Ye., P., et al., Method and system based on pulsed neutron generator for fissile material detection in luggage, Nucl. Instr. Meth. B 213 (2004) 439-444.
[4] SCHULTZ, F., J., et al., A feasibility study for a linac-based transuranic waste characterization system, J. Radioanal. Nucl. Chem., Articles, 193 (1995) 369-375.
[5] PIKSAIKIN, W., M., et al., Method and setup for measuring trace levels of heavy fissionable elements using delayed neutron counting, J. Nucl. Mat. Management 28 (2000), No. 3, 40-43.
[6] CHAKLOV, V., L., et al., Photoneutron source based on a compact 10 MeV betatron, Nucl. Instr. Meth. A 422 (1999) 5-9.
[7] CIERJACKS, S., SMITH, A. B., Neutron sources: Present practice and future potential, Nucl. Data for Sci. and Techn. (Proc. Int. Conf. Mito, 1988, IGARASI, S., Ed.), JAERI, Saikon (1988) 325-336.
[8] FRANKS, L., A., et al., High sensitivity transuranic waste assay by simultaneous photon and thermal neutron interrogation using an electron linear accelerator, Nucl. Instr. Meth. 193 (1982) 571-576.
[9] LYOUSSI, A., et al., Transuranic waste detection by photon interrogation and on-line delayed neutron counting, Nucl. Instr. Meth. B 160 (2000) 280-289.
[10] BOWMAN, C., D., Efficient neutron production using low-energy electron beams, Nucl. Sci. Eng. 75 (1980) 12-15.
[11] COCEVA, C., et al., Characteristics of pulsed neutron sources obtained with a low -energy electron accelerator Nucl. Data for Sci. and Techn. (Proc. Int. Conf. Mito, 1988, IGARASI,.S., Ed.), JAERI, Saikon (1988) 363-366.
[12] AUDITORE, L., et al., Study of a 5 MeV electron linac based neutron source, Nucl. Instr. Meth. B 229 (2005) 137-143.
[13] KEEPIN, G. R., et al., Delayed neutrons from fissionable isotopes of Uranium, Plutonium, and Thorium, Phys. Rev. 107 (1957) 1044-1049.
[14] TAM, N. C., et al., Monitoring bremsstrahlung from 4 MeV electrons by nuclear excitation and Al_2O_3 thermoluminescence dosimeters, Rad. Prot. Dosimetry 98 (2002) 401-406.
[15] BAGI, J., et al., Assessment of the Pu content of Pu-Be neutron sources, Nucl. Instr. Meth. B 222 (2004) 242-248.